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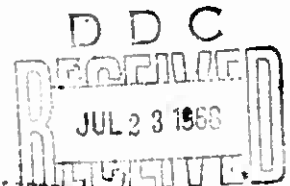
COMBUSTION OF SOLID PROPELLANTS AND LOW
FREQUENCY COMBUSTION INSTABILITY
PROGRESS REPORT 1 APRIL-30 SEPTEMBER 1967

by

Aerothermochemistry Division

Research Department

ABSTRACT. This semiannual report summarizes studies of ammonium perchlorate decomposition and deflagration, behavior of aluminum in the combustion zone of propellants, and oscillatory combustion of propellants in several types of experimental arrangements.



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FOREWORD

This report summarizes progress on a continuing program sponsored by the Office of Advanced Research and Technology of the National Aeronautics and Space Administration (NASA) under NASA Work Order 6030 during the period April through September, 1967. The preceding report on this work was NOTS TP 4244.

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NOMENCLATURE

- A Nondimensional parameter, $\frac{E_s}{RT_s} \left(1 - \frac{T_o}{T_s} \right)$
- A' Nondimensional parameter, $\left(\frac{E_s}{RT_s} - 1/2 \right) \left(1 - \frac{T_o}{T_s} \right)$
- A_b Area of burning surface
- A_t Throat area of sonic nozzle
- a Acoustic wave velocity
- B_{DB} Nondimensional parameter (see Eq. 4.9)
- B_{HFC} Nondimensional parameter (see Eq. 4.10)
- C_s, C_p Specific heat of solid and gas, respectively
- c* Characteristic velocity of exhaust gases
- D Diameter
- E_s, E_f Activation energy of the solid and the flame, respectively
- j Temperature sensitivity of propellant, $\frac{T_o}{r} \frac{\partial r}{\partial T_o}$
- K A constant, ≤ 1
- L Length of T-burner
- L* Characteristic length of chamber (free volume divided by nozzle throat area)
- ℓ_s Distance between entropy extremes
- m Mass burning rate

n	Pressure exponent in steady state burning rate law, $\bar{r} = bp^{-n}$
p	Pressure
\bar{p}	Mean or time-average pressure
p'	Pressure perturbation
p_r	Reference plane pressure
R	Gas constant
$\text{Re}()$	Real part of a complex expression
r'	Burning rate perturbation
s	Entropy
s_o	Reference entropy
T	Temperature
\bar{T}	Mean or time average temperature
T'	Temperature perturbation
T_f	Adiabatic flame temperature
T_o	Initial temperature of the propellant
T_r	Reference plane temperature
T_s	Surface temperature of burning propellant
T	Period of acoustic oscillation
V	Volume
\bar{V}	Velocity amplitude
v	Velocity perturbation
\bar{v}	Mean or time-average velocity
v_g	Mean gas velocity
X	Displacement
Y	Acoustic admittance

- α Exponential growth constant
- α_t Thermal diffusivity of solid
- c Ratio of maximum pressure perturbation to mean pressure
- λ Characteristic root of the solid phase energy equation (see Eq. 4.7 and wavelength in Section 5)
- μ/c Ratio of mass perturbation to pressure perturbation. Also designated the propellant response function.
- ρ Density
- u/c Ratio of density perturbation to pressure perturbation
- τ Characteristic time to burn through a particle
- τ_{ch} Residence time of the chamber
- τ_{tw} Thermal wave relaxation time, $\frac{\alpha_t}{r^2}$
- ϕ Phase of combustion relative to pressure
- ψ Phase between pressure and emergence of element of gas from the combustion zone
- Ω Non-dimensional frequency, $\frac{\alpha_t \omega}{r^2}$
- ω Frequency in radians/second

SUPERSCRIPITS

- Bar over a symbol (e.g., \bar{r}) refers to a time averaged value

SUBSCRIPTS

- b Burning surface
- g Gas
- o Total
- c Chamber

1. INTRODUCTION

1.1. CONCEPT OF STUDIES

The research work described in this report is part of a continuing program (Ref. 1) of investigation of the processes involved in low frequency acoustic and nonacoustic type oscillatory behavior in solid propellant rocket motors and in various aspects of steady-state combustion and ingredient decomposition believed to be related to solid propellant combustion instability. The present program was initiated at a time when there were two concurrent trends in the development of solid propellant rocket motors. The first of these was the development of very large solid propellant booster rockets, whose dimensions were such as to permit oscillatory behavior in acoustic modes at frequencies down to about 10 cps. The second was the growing interest in motors intended for space applications which possessed very high volumetric loading density and were designed to operate at relatively low pressures; both of these features are important for favorable mass ratios required for upper stage applications. Some motors of this type were observed to exhibit oscillatory behavior in a low frequency, nonacoustic mode, particularly in the frequency range of 10 to 200 cps (Ref. 2-4). Considering the extremely high cost of individual firings of the very large rocket motors under development and the cost of trial and error corrections should low frequency combustion instability occur, it was advantageous to develop laboratory scale tests which would permit qualitative evaluation of the susceptibility of propellants to low frequency combustion instability and to determine those attributes of the combustion responsible for instability.

A review of the general character of solid propellant combustion and of oscillatory combustion were presented in the previous report (Ref. 1); that review should be considered as background information for the work reported here.

1.2. PLAN OF THE REPORT

This report summarizes work that has been done on this program from April through September, 1967. Each section deals with a specific aspect of the program and in some cases is complete in itself. Although the sections are independent, they are mutually complementary; results in one area often suggest investigations that should be pursued in others.

Section 2 describes the continued investigations of decomposition and deflagration of large single ammonium perchlorate (AP) crystals. The

work included investigations with MnO_4^- -doped crystals and combustion of crystals in low pressure methane.

Section 3 reports the improvements in the apparatus and techniques for photographing propellant samples in a window bomb and also the progress in understanding the detailed aspects of aluminum accumulation in "real propellants".

Section 4 covers the work, both experimental and theoretical, on non-acoustic combustion instability. Experimental data was reasonably well correlated by the models of Denison and Baum and of Hart, Farrell, and Cantrell. A phenomenological concept is presented which partially explains the deviation of experimental data for several bimodal oxidizer propellants from the one-dimensional thermal theory.

Section 5 deals with the work on low frequency acoustic instability with particular emphasis on the dynamics of the combustion zone as inferred from streak photographs and gas temperature measurements taken during oscillatory combustion.

Section 6 is a brief summary of the more significant observations made during this reporting period and their relation to previous work on the program.

Appendix A consists of burning rate data and tabular experimental data obtained on nonacoustic combustion instability. Appendix B presents the results of testing some of the NASA big booster candidate propellants in both the acoustic and nonacoustic laboratory burners and discusses their significance.

2. AMMONIUM PERCHLORATE DECOMPOSITION AND DEFLAGRATION

2.1. INTRODUCTION

Ammonium perchlorate (AP) is the oxidizer and principal ingredient in most modern composite propellants and its behavior in the combustion wave is an essential aspect of the combustion as a whole. The knowledge of the major aspects of the low temperature decomposition and high temperature deflagration of AP was summarized in the previous report (Ref. 1). Work has continued in both these areas of AP behavior. In the thermal decomposition area, single crystals of pure AP were partially decomposed and subsequently studied with a scanning electron microscope and the interfacial decomposition of MnO_4^- -doped AP crystals was investigated. In the area of AP deflagration, further studies were conducted to elucidate the extent of the occurrence of a melt on the surface of pure AP crystals undergoing self-deflagration. Also, deflagration studies have been started on doped AP single crystals.

2.2. LOW TEMPERATURE DECOMPOSITION EXPERIMENTS¹

2.2.1. Surface Structure of Partially Decomposed AP Crystals

To gain additional insight into the low temperature decomposition of AP, a series of experiments were undertaken to systematically study the surface features of partially decomposed AP crystals.² The isothermal decomposition of single crystals of AP was monitored with a hot stage microscope and stopped at a predetermined stage. The samples were then vacuum coated with an Au/Pd alloy and kept under vacuum until they could be photographed with the scanning electron microscope (SEM). Several series of SEM photographs are shown of AP samples decomposed under various conditions. Each figure contains pictures of four different magnifications to allow the study of details as well as bigger areas of a crystal surface. Rectangular-shaped holes are characteristic for decomposition

¹ This work was supported by Naval Ordnance Systems Command ORO-033 129/200 1/R001-06-01, Problem Assignment #5 and is reported here for continuity.

² Both the pure and doped AP crystals used in the studies in this section were grown by Dr. W. R. McBride of the Research Department, Michelson Laboratory, NWC, China Lake, California. A description of the techniques is given in Ref. 1.

sites on *m*-faces whereas rhombohedral-shaped holes are characteristic for the decomposition sites on *c*-faces. Figure 2.1 shows the *m*-face of a crystal which was decomposed isothermally at 226°C for 40 minutes at an air pressure of 20 mm Hg. The characteristic feature is the rectangular structure of the holes which make up the decomposition nucleus. The holes have the shape of the *m*-face with edges running parallel to the edges of the *m*-face. They are arranged in a disk-like pattern and eventually develop into a porous residue. Furthermore, the decomposition seems to occur in layers with the size of the holes becoming smaller and smaller with depth. There is a second type of decomposition site which generally does not develop into growing nuclei composed of holes. They appear immediately after reaching the decomposition temperature and were not observed under atmospheric pressure.

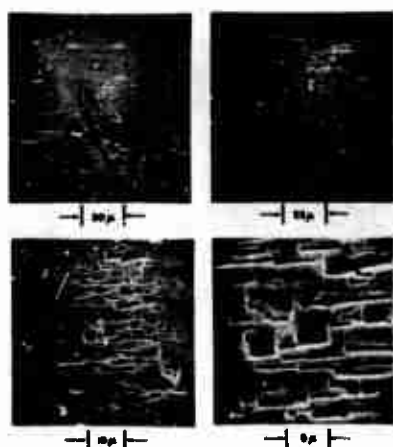


FIG. 2.1. Decomposition Site on *m*-Face of Pure AP After 40 Minutes Decomposition at 226°C and 20mm Hg.

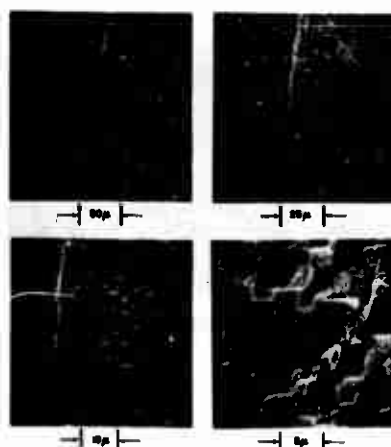


FIG. 2.2. Decomposition Site on *c*-Face of Pure AP After 60 Minutes Decomposition at 226°C and 20mm Hg.

Figure 2.2 shows part of a *c*-face on a crystal which was decomposed for 60 minutes at 226°C and 20 mm Hg of air. Analogous to *m*-faces there are two types of decomposition sites which have the shape of rhombohedrons. One type consists of holes arranged in a streak-like pattern. The streaks grow faster along the *b*-axis than along the *a*-axis and, after overlapping, constitute the porous residue layer. The other type comprises very flat rhombohedral holes which are spread all over the *c*-faces and appear only under low pressure. Both the streaks and the long diagonals of both types of decomposition sites are parallel to the *b*-axis of the crystal. If the decomposition is carried on for a longer time the

surface becomes less regular, although some of the rectangular structure is still preserved, as can be seen in Fig. 2.3, which shows part of an *m*-face after 140 minutes decomposition at 226°C and a pressure of 20 mm Hg air. There is a very different type of structure at the very top of the decomposition site in Fig. 2.3. It appears to consist of clusters of very small AP particles. The clusters may be products of a complex dissociation-desorption-adsorption process or may be residues of advanced sublimation of the porous layer.

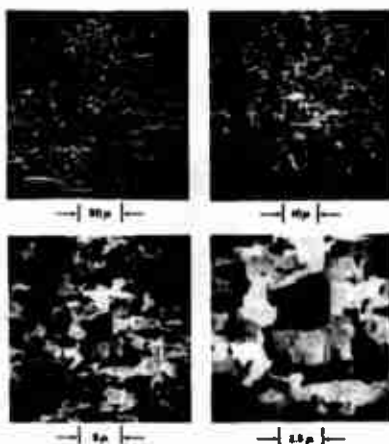


FIG. 2.3. Decomposition Site on *m*-Face of Pure AP After 140 Minutes Decomposition at 226°C and 20 mm Hg.

The decomposition under atmospheric pressure does not change the general appearance of the decomposition sites. Figure 2.4 shows part of an *m*-face after 90 minutes decomposition at 226°C and Fig. 2.5 part of a *c*-face after 120 minutes decomposition.

However, some differences from vacuum decomposition can be seen. First, the size of the holes is smaller although the crystals decomposed about twice as long as under vacuum. This is probably due to a suppression of sublimation at atmospheric pressure. Second, there is a great number of fine cracks not visible on vacuum decomposed samples, and third, there are spots of elevated surface. These spots are concentrated at the rim of an *m*-face decomposition site whereas they are abundant over the whole streak on a *c*-face.

As a by-product of the decomposition at both 20 mm Hg and at atmospheric pressure a condensate was obtained on a cool (about 50°C) quartz plate. Figure 2.6 shows a SEM photograph of the condensate obtained during the decomposition of an AP crystal at one atmosphere of air. The condensate was in the early stage of development. An x-ray powder pattern showed the condensate to be pure AP. This indicates that there was

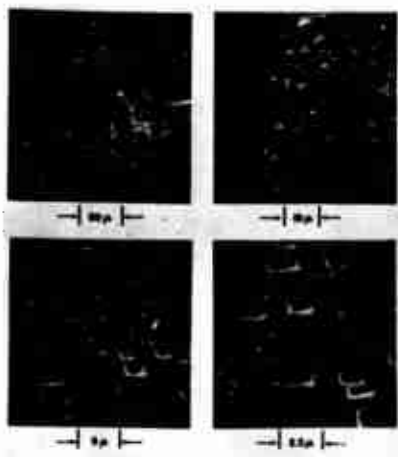


FIG. 2.4. Decomposition Site on *m*-Face of Pure AP After 90 Minutes Decomposition at 226°C and Atmospheric Pressure.

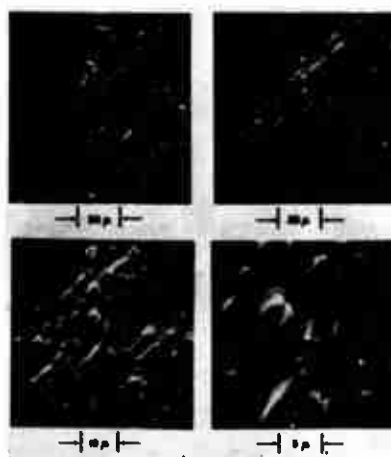


FIG. 2.5. Decomposition Site on *c*-Face of Pure AP After 120 Minutes Decomposition at 226°C and Atmospheric Pressure.

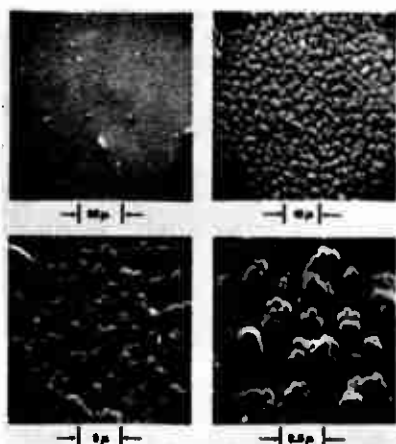


FIG. 2.6. Condensate of Pure AP on Quartz Disk.

an appreciable sublimation of AP even at atmospheric pressure. No condensate is obtained if the whole reaction vessel (volume) is kept at decomposition temperature. In that case, the part of AP which sublimes subsequently decomposes in the gas phase or, heterogeneously, on the hot surface (walls) of the oven. In view of continuing discussions of condensed and/or gas phase reactions during the decomposition of AP, and in light of the above observations, an effort should be made to obtain data simultaneously for reactions occurring in the condensed phase and away from the condensed phase of decomposing AP.

2.2.2. Interfacial Decomposition of Doped AP Crystals

Work in this area as reported in Ref. 1 has been expanded to include two additional concentrations of MnO_4^- in the doped AP single crystals. The procedure is described and results for pure AP crystals were reported in Ref. 1.

Single AP crystals containing 0.009 weight percent, 0.04 weight percent, and 0.11 weight percent MnO_4^- were decomposed isothermally under atmospheric pressure. The advance of the decomposition interface as a function of time was obtained by cleaving the crystals and measuring the thickness of the residue layers at different stages of decomposition. The total weight loss after a complete partial decomposition was between 35 and 40% of the original weight as compared with 35% for pure AP and about 30% measured by Bircumshaw and Newman (Ref. 5).

As in the case of pure AP, it was found that the decomposition interface advanced linearly with time into the crystal. Figures 2.7 through 2.12 show data of *m*- and *c*-face decomposition. The decomposition parallel to *c*-faces is about 10-20% slower than the decomposition parallel to *m*-faces. With the rate values evaluated from Fig. 2.7 through 2.12 one can calculate the fractional decomposition at a given time for crystals which were cleaved to the shape of a parallelepiped. Of more interest, however, are the kinetic parameters of the isothermal decomposition which can be obtained from Arrhenius plots of rates versus $1/T$. Figure 2.13 shows the diagrams for *m*-face decomposition and Fig. 2.14 for *c*-face decomposition. The kinetic parameters calculated from these figures are listed in Table 2.1. Values reported for pure AP single crystals (Ref. 1) were 6.5×10^3 cm/sec for the preexponential factor and 21.9 kcal/mole for the activation energy (average values for *m*- and *c*-faces). All of the doped AP had preexponential factors and activation energies smaller than the values for pure AP. Accordingly, at higher temperatures and under the assumption that the decomposition mechanism stays the same, one would expect smaller burning rates for doped AP because the preexponential term becomes more and more dominant and the Arrhenius plots intersect at relatively low temperatures. This was in fact observed (Section 2.3.2), but it does not infer that the combustion of AP obeys the same or similar mechanisms as the low temperature decomposition. This would be a gross simplification of a very complex process.

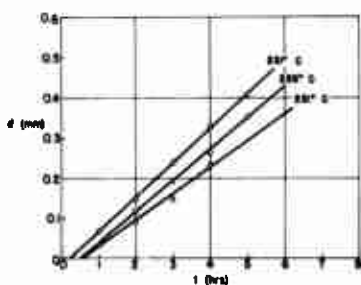


FIG. 2.7. 0.009 Wt.% MnO_4^- .
m-Face Decomposition.

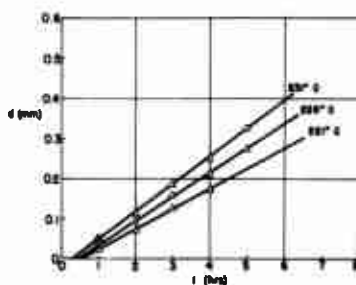


FIG. 2.8. 0.009 Wt.% MnO_4^- .
c-Face Decomposition.

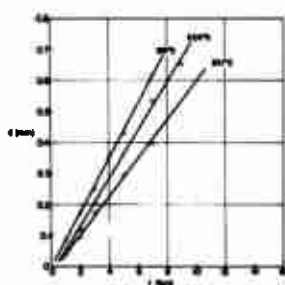


FIG. 2.9. 0.04 Wt.% MnO_4^- .
m-Face Decomposition.

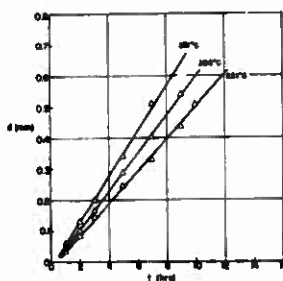


FIG. 2.10. 0.04 Wt.% MnO_4^- .
c-Face Decomposition.

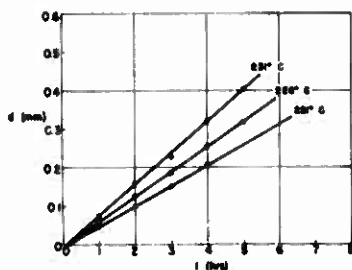


FIG. 2.11. 0.11 Wt.% MnO_4^- .
m-Face Decomposition.

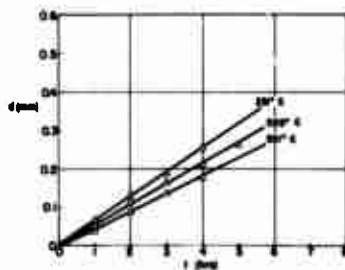


FIG. 2.12. 0.11 Wt.% MnO_4^- .
c-Face Decomposition

FIG. 2.7-2.12. Penetration of the Decomposition Interface Into NH_4MnO_4 -Doped Ammonium Perchlorate Single Crystals. d = distance from crystal surface to interface.

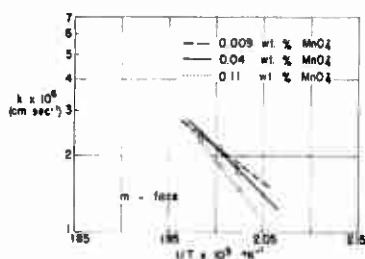


FIG. 2.13. Arrhenius Plot for NH_4MnO_4 -Doped AP. m-Face Decomposition.

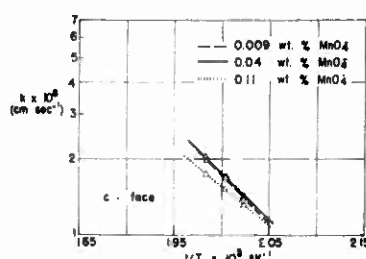


FIG. 2.14. Arrhenius Plot for NH_4MnO_4 -Doped AP. c-Face Decomposition.

TABLE 2.1.

KINETIC PARAMETERS FOR THE DECOMPOSITION OF NH_4MnO_4 -DOPED AMMONIUM PERCHLORATE SINGLE CRYSTALS

SAMPLE	PREEXP. FACTOR (cm/sec)		ACTIVATION ENERGY kcal/mole	
	m-face	c-face	m-face	c-face
AP	0.9	62.0	12.9	17.3
+ 0.009 WT. % MnO_4^-				
AP	47.3	39.0	16.8	16.7
+ 0.04 WT. % MnO_4^-				
AP	4180.0	2.5	21.4	14.2
+ 0.11 WT. % MnO_4^-				

A theoretical interpretation of the results has not been tried yet in view of the uncertainty concerning the nucleation rate and rate of nuclei growth. In preliminary experiments using a hot stage optical microscope it was found that the nucleation rate of doped AP is much higher than that of pure AP. In addition, the number of nucleation sites is orders of magnitude greater for doped than for undoped AP if both are compared after the same decomposition time. This may affect the preexponential factors as well as the activation energies and a careful study of nucleation and nuclei growth, both for pure and doped AP, will be necessary to give an interpretation of the variety of kinetic parameters available.

2.3. SINGLE CRYSTAL DEFLAGRATION STUDIES

2.3.1. Combustion In Reactive Atmospheres

In earlier work (Ref. 1, 6 and 7) evidence was reported which indicated that a thin layer of melt was present on the surface of pure AP crystals undergoing self-sustained deflagration. The studies have been extended to determine the extent of occurrence of the melt and its importance to the deflagration process. The use of reactive atmospheres such as methane and ammonia have allowed studies to be made at reduced pressures corresponding to regression rates intermediate between those of self-deflagration and low temperature decomposition.

Single crystals of pure AP and crystals doped with various concentrations of NH_4MnO_4 and KClO_4 were grown in the apparatus described in Ref. 1. Figure 2.15 shows crystals typical of those used in the present work. Samples were burned in the flushed window bomb described in Section 3.2. High speed motion pictures were taken to study the qualitative aspects of combustion and to determine the burning rates. For photographic purposes pure AP and K^+ -doped crystals were illuminated by a 2,500-watt xenon lamp. The purple MnO_4^- -doped AP crystals were photographed with transmitted, low intensity polarized light to prevent heating the crystals by optical radiation absorption.

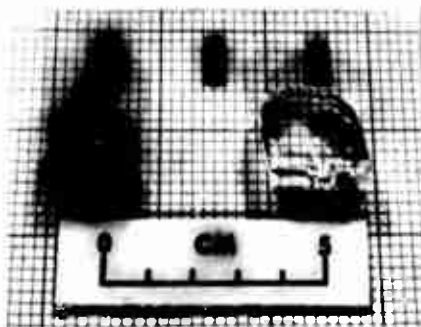


FIG. 2.15. Single Crystals Typical of Those Used in the Present Work. At left is a MnO_4^- -doped crystal which has deep purple color and slightly modified growth habit. At right is pure AP crystal and above small, cleaved segments of MnO_4^- -doped AP in various concentrations.

High speed motion pictures of the deflagration in methane at pressures between atmospheric and 100 psia were of particular interest. Under these conditions a variety of surface heating conditions and, hence, regression rates was possible. Figure 2.16 shows a pure AP crystal burning at 15 psia in methane. At this pressure the surface regressed with a pronounced coning due to flame spread down the sides of the crystal; the upper portion of the coned surface had a regression rate of approximately 0.03 in/sec. The surface had a frothy, translucent appearance. Quenched samples at these lower pressures were obtained by rapidly flooding the combustion bomb with nitrogen. Figure 2.17 is a scanning electron microscope (SEM) photograph of the surface of a



FIG. 2.16. Single Frame of Motion Picture of Pure AP Burning in Methane Atmosphere at 15 psia. Crystal approximately 0.5 cm wide.

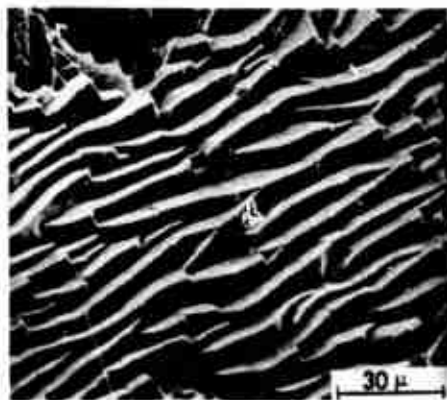


FIG. 2.17. Scanning Electron Microscope Photomicrograph of the Burned Surface of Pure AP Quenched from Combustion in 15 psia methane.

quenched sample that was burned in this intermediate regression rate domain. The surface structure that results from this mode of combustion gives the appearance of a crystal structure but differs from the structure of an "undisturbed" AP crystal. Figure 2.18 shows a pure AP crystal burning at 65 psia. At this pressure the surface was clear and transparent and resembled melting ice. Occasional boiling activity could be clearly seen on the surface. At 100 psia in methane movies taken at a magnification of 4X of AP crystals approximately 250 micron thick showed a definite "flowing" motion associated with portions of the burning surface as well as patches of rapidly boiling "froth". It was not possible to quench samples under these conditions of higher pressure.



FIG. 2.18. Single Frame of Motion Picture of Pure AP Burning in Methane Atmosphere at 65 psia. Crystal approximately 0.5 cm wide.

The K^+ -doped AP, as observed in the movies, behaved essentially like pure AP when burning in methane except that the tendency for surface boiling was more pronounced and easily observed.

In low pressure (15 to 50 psia) methane the MnO_4^- -doped crystals were observed to burn with a white porous material covering the regressing surface. Figure 2.19 is a SEM photograph of the surface of a MnO_4^- -doped crystal quenched at low pressure. This structure is very similar to the water vapor "rejuvenated" low temperature decomposition residue shown in Fig. 2.20. At higher pressure (200 psia) the porous material appeared to undergo a cyclic accumulation-rapid consumption process which gave a strange pulsating character to the deflagration process.

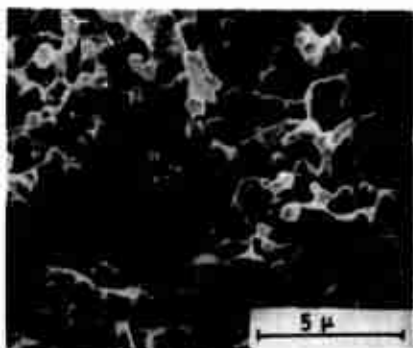


FIG. 2.19. Scanning Electron Microscope Photomicrograph of the Burned Surface of MnO_4^- -Doped AP Quenched from Combustion in 15 psia Methane.

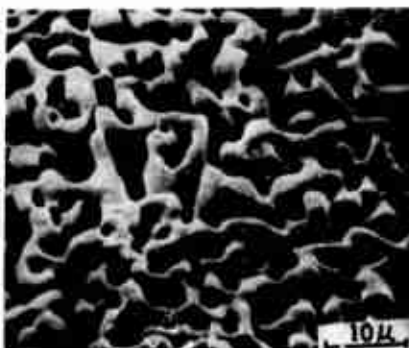


FIG. 2.20. Scanning Electron Microscope Photomicrograph of the Residue of Partially Decomposed Pure AP (at $220^\circ C$) Which has Been Rejuvenated by Exposure to Water Vapor.

Low speed motion pictures (24 to 100 fps) were taken utilizing the self-illumination of the AP decomposition flame. At atmospheric pressure in methane a pink-orange flame approximately 175 microns thick was observed (by visual observation the flame color was the same as was observed with single crystals burning in nitrogen at elevated pressure). The decomposition flame appeared to commence right at the condensed phase surface.

In addition to the samples burned in methane, several pure AP crystals were burned in an atmosphere of ammonia at atmospheric pressure. The combustion behavior was essentially the same as in methane atmospheres. It was not possible to burn samples at elevated pressure in ammonia since the crystals rapidly dissolved in the ammonia.

2.3.2. Deflagration at High Pressure

Additional measurements were made of the deflagration rates for single crystals. The rates were measured from high speed motion pictures of the combustion and are shown in Fig. 2.21. The previously reported AP burning rate data (Ref. 6) were repeated and verified with larger cross-section (0.5 cm on a side) crystals.

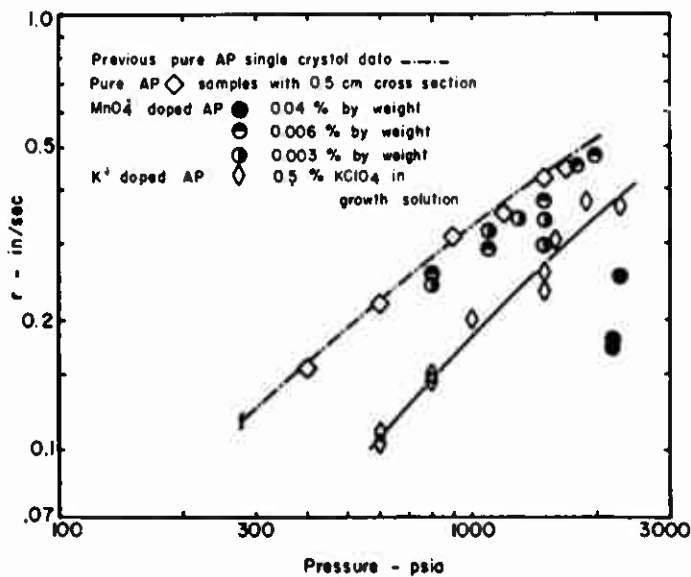


FIG. 2.21. Deflagration Rate of Pure and Doped AP Single Crystals.

The burning rates for the single AP crystals doped with K^+ and three concentrations of MnO_4^- exhibited one consistent feature even though there was inconsistency in some of the rate-concentration data; that feature was that the burning rates for the doped crystals were always below the pure AP rate over the pressure range tested. In addition, the K^+ -doped AP had a pressure exponent of approximately 1.0 as compared to approximately 0.77 for pure AP. Motion pictures of this material undergoing deflagration showed definite melting and boiling on the surface with occasional ejection of molten material (presumed KCl) from the surface.

In the case of the MnO_4^- -doped crystals, the burning rate data at the lowest concentrations was too scattered to warrant drawing conclusions concerning rate-concentration trends. In addition, it was difficult to precisely measure the MnO_4^- concentration. The highest (0.04%) concentration MnO_4^- -doped AP which would undergo self-deflagration displayed a much lower burning rate so it is presumed that increasing MnO_4^- concentration results in lowering the single crystal deflagration rate. Light transmission through the gaseous combustion products was observed to be much higher for the MnO_4^- -doped AP than for the pure AP. Figure 2.22 shows a comparison of doped versus pure AP photographed under the same test conditions with transmitted illumination by polarized light.

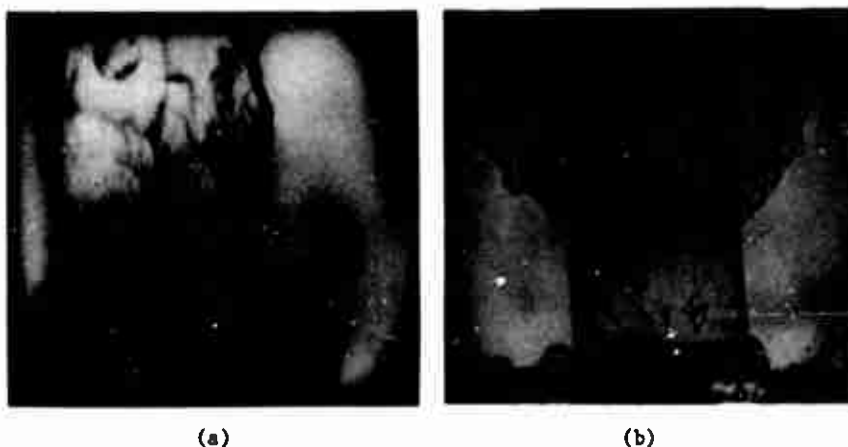


FIG. 2.22. Single Frame of Motion Picture of (a) Pure AP and (b) AP Doped With 0.006% MnO_4^- . Both samples deflagrating at 800 psia, photographed with transmitted, polarized illumination. Note difference in optical density of combustion products.

The low pressure deflagration limit for all of the doped crystals was much higher than that of the pure material (275 psia, Ref. 6) with MnO_4 being apparently more effective in raising the limit than K^+ . Friedman, et al (Ref. 9), also observed that $\text{NaMnO}_4 \cdot 3\text{H}_2\text{O}$ and MnO_2 raised the deflagration limit of pressed AP pellets. The low pressure deflagration limits in the present work were found to be approximately 600 psia for the 0.5% KClO_4 -doped AP and 800 psia for the 0.003 to 0.006% MnO_4 -doped AP. The AP doped with 0.04% MnO_4 could not be made to deflagrate below about 2000 psia.

2.3.3. Interpretation and Discussion

The work here and in Ref. 1 has revealed several qualitative aspects of combustion that were previously unknown, or were matters of speculation. The most important aspect is the decisive evidence for a frothy liquid on the surface of the burning AP. The exact chemical nature of the liquid is not known, although x-ray diffraction tests on residues indicate only AP. It is not surprising that a liquid layer should be present at the high surface heating rates associated with self-deflagration of crystalline AP. The temperature gradient resulting from this heat flux is of such magnitude that it could raise the temperature of the subsurface AP to its melting point in less time than it would take to gasify the material. Thus, a melting point would be encountered which would not be observed at lower heating rates, i.e., such as associated with differential thermal analysis, where the sample would gasify before the melting point was reached.

The results suggest that the melt plays an important role in self-deflagration of the AP. There has been continuing difficulty in reconciling the amount of heat required for the initial decomposition of AP with the amount of heat that could be supplied by the AP flame, because the heat balance dictates a flame standing only a few mean free paths from the surface (assuming a one-dimensional regressing surface). On the other hand, a frothy melt on the surface provides opportunity for both heterogeneous and gas phase reactions in the froth, with a very large area for heat transfer. Thus, it seems plausible to assume that the liquid froth is an essential feature of the self-deflagration of AP, and that the low pressure deflagration limit corresponds to attainment of heating rates so low that surface material gasifies without reaching a melting temperature. Under these conditions, a major portion of the exothermic reactions would occur only in the gas phase flame, which apparently does not have any equilibrium position satisfying heat balance requirements.

Microscopic examination of quenched samples indicate no optically detectable changes in the AP below the melt region other than the phase transition region. This implies that there is probably no significant contribution to the heat release during deflagration by solid phase reactions. This is not really surprising because the decomposition reactions of solids include an induction period followed by a growth and

decay period. The induction period may be long or at least comparable to the time it takes for the very thin thermal wave associated with combustion to traverse a given layer of material. Ammonium perchlorate does decompose in a combustion wave, and on a molecular scale the actual decomposition mechanism may be the same as for the "low temperature" thermal decomposition, e.g., a proton transfer. The kinetics of the overall process are expected to be different if they occur in a melt as opposed to a crystalline solid.

It seems reasonable to assume then, that alteration of the thermal stability of the surface melt or suppressing its formation would produce a corresponding change in the deflagration behavior of AP. Earlier work (Ref. 9) showed that a rapidly heated mixture of granular AP and lithium fluoride produced a residue mixture of AP-lithium perchlorate. Markowitz and Harris (Ref. 10) have shown that the decomposition rate of AP in solution with lithium perchlorate is lower than that of pure AP at corresponding temperatures. It is possible that a similar process may occur on the surface of deflagrating crystals doped with the K^+ ion resulting in the formation of an AP-potassium perchlorate solution. There is also an accumulation of liquid balls (probably KCl , a stable product of $KClO_4$ decomposition) on the burning surface of K^+ doped crystals which may affect the deflagration rate. In the case of MnO_4^- -doped crystals both the deflagration limit and the burning rate were influenced tremendously by small concentrations of this ion. The pressure deflagration limit is increased accordingly along with a reduced burning rate.

2.4. SUMMARY

The present work (including Ref. 1) has revealed rather decisively that the physical character of the surface residue of AP is significantly dependent on the imposed heating rate. Under conditions of isothermal decomposition the AP apparently sublimates and there is no evidence of a surface melt. Under conditions of rapid heating, a surface melt is observed and in the case of self-deflagration the surface melt is frothy and may contribute significantly to the heat release necessary to sustain deflagration. Also, the substitution of foreign ions in the AP crystal structure in small quantities reduced the activation energy of the isothermal decomposition and also altered the combustion behavior of the crystal either by altering the thermal stability of the surface melt and/or the gas phase kinetics.

3. BEHAVIOR OF ALUMINUM IN COMPOSITE PROPELLANTS

Certain aspects of aluminum agglomeration in the combustion zone of composite propellants were described in detail in the previous progress report (Ref. 1). During this reporting period, emphasis has been placed on investigating the mechanism of agglomeration. In particular, the photographic technique has been improved to increase the resolution of the high speed motion pictures, and quenched propellant samples have been examined in detail with a scanning electron microscope (SEM) to investigate the early (preignition) phases of aluminum agglomeration.

3.1. EXPERIMENTAL TECHNIQUE DEVELOPMENTS

3.1.1. Cinephotomicrography Improvements

The basic arrangement for high speed motion picture photography with magnification has been described in Ref. 1. However, improvements have been made in each of the three major components of the experimental setup.

The improved arrangement is shown in Fig. 3.1. The original Fastax camera was replaced by a Hycam 16 mm high speed motion picture camera, Model K3004E. This camera provides more uniform exposure over the entire film frame, more versatility in speed selection and, due to its internal optics, provides a somewhat larger magnification than the Fastax. The original xenon arc external illumination arrangement was replaced with a Strong X-16 xenon arc assembly which includes a 2,500 watt xenon arc lamp, dichroic main mirror and external focusing controls. This assembly permits better control of external illumination and can supply considerably more intensity, if desired, than the previous arrangement. The greatest improvement has been made in the window bomb itself. The new bomb provides complete flushing of the interior without recirculation, permits more versatility in the mounting of propellant samples (Fig. 3.2), and provides better mechanical support for the fused quartz windows.

Figure 3.3 illustrates the details of combustion that can now be resolved with the improvements in the experimental arrangement. In the first picture, an accumulation of several hundred 15 μ aluminum particles have melted together to form a sphere of molten aluminum, as indicated by the highlight on the sphere. Some unmelted material, including several original aluminum particles, is visible on the left side of the agglomerate. In the second picture (one millisecond later) most of the remaining material on the agglomerate has melted except for the crusty,

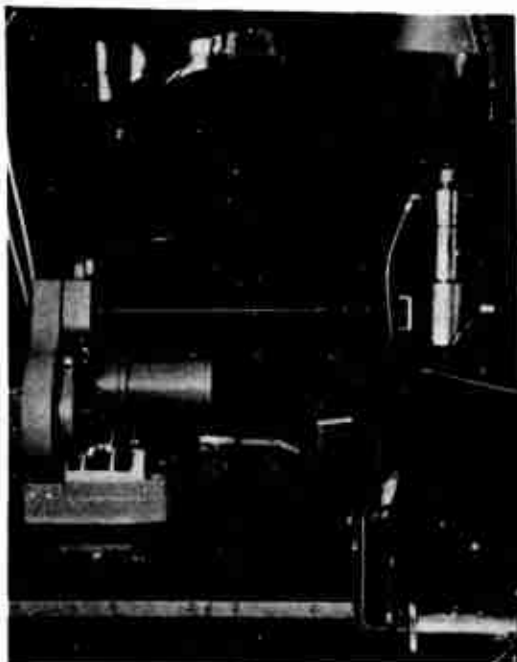


FIG. 3.1. Photographic Arrangement.

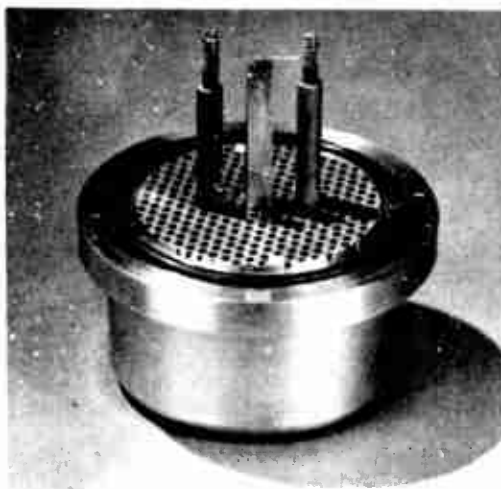


FIG. 3.2. Propellant Sample Holder.



t = 1 MSEC



t = 1 MSEC



t = 5 MSEC



t = 8 MSEC



FIG. 3.3 Selected frames from high speed motion picture of composite propellant containing 18% aluminum burning at 100 psi.

irregular appearing aluminum oxide surface. The aluminum oxide "smoke" indicates the developing reaction region of the aluminum combustion. At five milliseconds the droplet shows a well-developed combustion envelope which flows outward with the propellant combustion products. In the final picture the spherical droplet appears to be burning vigorously and highlights from the liquid surface are visible through the combustion envelope. The disc centered on the sphere in this picture is a luminous cap which probably contains most of the preignition oxide of the original aluminum particles. Such oxide caps are readily discernible in the photographs in Ref. 1 (Fig. 3.3). The final picture also shows the heterogeneity of the propellant surface underneath the aluminum agglomerate. The motion pictures show this area to be almost entirely "uncluttered" with pyrolysis residue. Assuming that the residue is a thermal insulator, energy from the gas phase could be more easily transferred to this "clean" spot and could result in a momentary increase in energy release in that localized area. This possibility along with the energy release following ignition of the aluminum agglomerate quite conceivably could play an important role in driving gas oscillations.

3.1.2. Propellant Quench Technique

Quenching propellants by rapid depressurization of the window bomb was performed earlier in this program (Ref. 11). High speed motion pictures taken during the quench indicated that the majority of aluminum in the later stage of agglomeration was removed from the surface during the rapid depressurization. In order to preserve the large accumulates during quench for later microscopic study, a new quench technique was employed. A freshly cut piece of propellant (approximately a 1/4 in. cube but with the top surface cut at a 30° angle to the horizontal) was placed on a clean metal plate and ignited by a hot Michrome wire in the pressurized bomb. As the burning surface approached the metal plate, the combustion was thermally quenched. A small portion of unburned propellant as well as aluminum particles in various stages of agglomeration remained on the plate. Plates of copper, aluminum, and stainless steel were evaluated as heat sinks; the stainless steel plates were most satisfactory in that a larger portion of unburned propellant remained after quench. Some residuums have been examined with a scanning electron microscope (SEM) and the results are discussed in later sections.

3.2. DETAILED ASPECTS OF ALUMINUM AGGLOMERATION

The preignition behavior of the aluminum in composite propellants has received increased attention during this reporting period and the more important processes are reasonably well understood. An exploratory experiment which yielded the greatest insight into the agglomeration mechanism was performed by dropping 100 μ spherical aluminum particles (in air) onto a sapphire disk heated by a torch to approximately 1,400°C (Ref. 12) and subsequently examining the particles using a scanning electron microscope (SEM). A series of resulting SEM photographs with

increasing magnification are shown in Fig. 3.4. The important feature shown in Fig. 3.4a is that two aluminum particles are rigidly connected by a "bridge" of material. This interaction of the particles occurred at an environmental temperature ($< 1,400^{\circ}\text{C}$) well below the melting point of aluminum oxide ($2,050^{\circ}\text{C}$), even though the original aluminum particles had an oxide coating. Examination at higher magnifications shows numerous ridges approximately $1/2\mu$ in width which resemble welding joints. These observations on the microscopic scale probably explain the details of aluminum agglomeration in propellants and are interpreted in the following manner. As the aluminum particles are heated (in this experiment by conduction from the $1,400^{\circ}\text{C}$ sapphire disk), the stresses produced by the differential thermal expansion of the aluminum and aluminum oxide caused the aluminum oxide covering to fracture. The particles in Fig. 3.4 were evidently heated to above the melting point of aluminum and hence, liquid aluminum started to flow through the cracks. The aluminum was rapidly oxidized, thus producing the "weld joint" appearance. The heat liberated by the aluminum oxidation conceivably could produce additional stresses and cracking if radiative heat losses, for instance, were not too great. As in Fig. 3.4, where the particles are in contact, a "bridge" can be formed by the cracking of oxide and flow of molten aluminum. In Fig. 3.4d it appears that additional "thermal cracking" occurred after the bridge was formed. This is indicated by the two ridges that run across the bridge.

It is felt that this phenomena is the first step in the agglomeration (or sintering together) process. Combustion residues obtained from thermal quenching (Section 3.1.2) were examined with the SEM for evidence of the "bridging" phenomena. Figure 3.5a is a series of SEM photographs of an agglomerate from a composite propellant containing 16% of 15μ diameter aluminum. The successively larger magnifications show the pattern of the aluminum flowing to form a large agglomerate. For comparison, Fig. 3.5b shows an analogous situation of agglomerate formation with 100μ aluminum particles rapidly heated to $1,400^{\circ}\text{C}$ on a sapphire disk in atmospheric air. The details of oxide cracking and healing are not at all evident in the agglomerate from the propellant. This agglomerate was formed in a more complicated environment and undoubtedly there are numerous pyrolysis and oxidation products present which could greatly modify the appearance of the agglomerate. However, efforts are continuing to conclusively demonstrate this "bridging" phenomena because of the importance of relating the metal particle behavior with behavior in propellants.

3.3. INTERPRETATION AND SUMMARY

A detailed, qualitative description of the behavior of aluminum in composite propellants can now be made based on (1) high speed motion pictures of aluminized propellants, especially the observations of a liquid phase on the propellant surface for the early stages of accumulation, (2) on interpretation of and deductions from the observations of

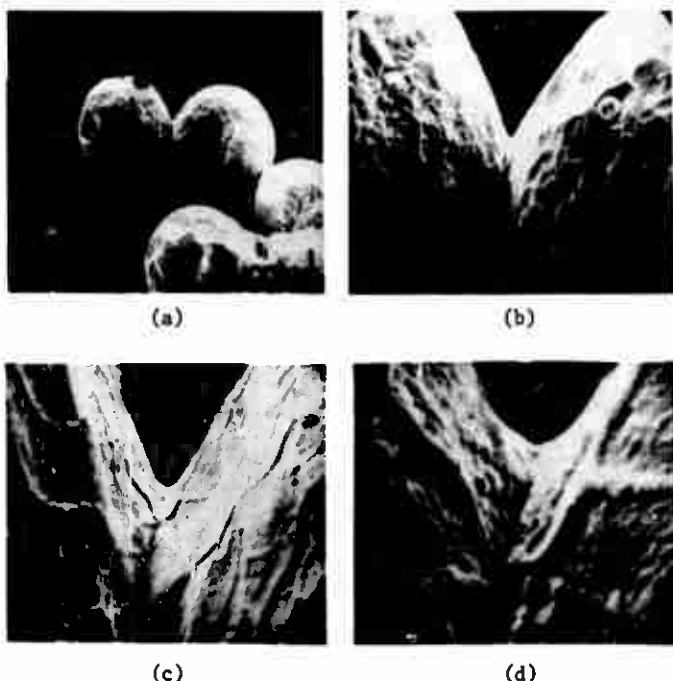


FIG. 3.4. SEM Photographs of "Bridges" Joining 100 μ Aluminum Particles. Aluminum particles were dropped onto sapphire plate heated to 1,400°C in air.

the behavior of rapidly heated single particles in Section 3.2 and similar, though less conclusive, studies of residual accumulates from propellant quench tests, and (3) on high speed motion pictures (Section 3.1.1 and Ref. 1 and 13) of burning propellants as exemplified by the four frames shown in Fig. 3.3.

The sequence of events in the agglomeration of aluminum is believed to be as follows. The original aluminum particles with thin oxide coatings are distributed in the binder of the propellant. As the combustion front moves through the propellant, the temperature of the aluminum particles begins to rise as the thermal wave proceeds. As the binder and the ammonium perchlorate pyrolyze, the aluminum recedes with the surface until it comes into contact with other aluminum particles. The decomposing surface of the propellant is liquid and thus the individual aluminum particles are readily retained on the surface. As the aluminum particles accumulate in the melt, they are heated by conduction to at least the local surface temperature (typically on the order of 400°C-700°C) and probably reach a somewhat higher temperature due to oxidative

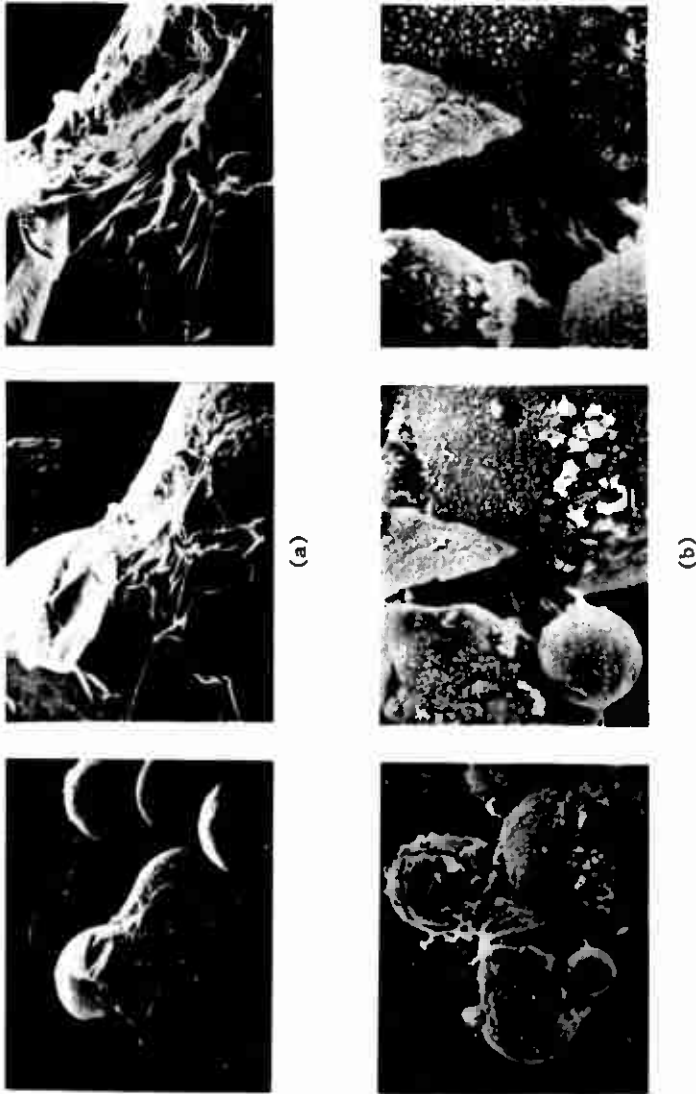


FIG. 3.5. SEM Photographs of Aluminum Agglomeration. (a) Particles dropped onto 1,400°C plate, (b) Quenched particles from composite propellant.

self-heating. In this situation with the aluminum particles in contact with one another and with the aluminum at a temperature above its melting point, the adjacent particles interact as described in Section 3.2. The molten aluminum flows through the thermal expansion cracks in the aluminum oxide shell and is probably quickly oxidized. When cracks roughly coincide with the contact points with adjacent particles, the flowing aluminum can "wet" the adjacent particle. Thus, the aluminum particles are joined together at their contact points by aluminum and aluminum oxide "bridges". The agglomerate as a whole can be described as numerous drops of liquid aluminum encapsulated in aluminum oxide shells. These drops and shells are linked together by oxide "bridges". Figure 3.4 represents this type of structure. As the binder and AP recede beyond this region of the aluminum agglomerate, the aluminum-aluminum oxide structure has enough strength to physically withstand the flow of binder and oxidizer gases away from the surface and, hence, remains on the surface until it ignites or is completely undermined and drifts away with the flow of gas from the surface.

In the case where the agglomerate is not undermined and removed, oxidative self-heating, oxide cracking and aluminum flow continues until the molten aluminum from one shell joins with aluminum from another and a large molten ball of aluminum has formed. The latter stage of the formation of a large molten ball of aluminum is shown in the "t" portion of Fig. 3.3. The next picture ($t + 1$ msec) shows the ball increasing in size to engulf the entire agglomerate contained in that particular binder "pocket". These two pictures also show that the aluminum oxide on the large molten ball is irregular in shape indicating that the oxide has not yet melted. In the $t + 5$ msec picture the oxide has melted (no sharp irregularities) and vigorous combustion of the aluminum has begun as indicated by the pronounced aluminum oxide smoke that has developed. The molten aluminum oxide cap characteristic of burning aluminum droplets is distinguishable in the $t + 8$ msec picture.

Future effort in this area will be directed at obtaining even more conclusive evidence on the aluminum sintering process and the transition to a large molten ball in experiments closely simulating the propellant combustion situation.

4. NONACOUSTIC INSTABILITY

4.1. INTRODUCTION

A general review of the status of nonacoustic combustion instability (NAI) in solid propellants was presented in the previous report (Ref. 1). The principal effort during this reporting period has been (1) to compare nonacoustic experimental results with one-dimensional theories and (2) to examine the effect of oxidizer particle size on NAI.

In the past, theoretical models describing the interaction of the pressure perturbations with the combustion zone have mainly been applied to acoustic instability (for example, Ref. 14-17). However, the combustion instability models that describe the dynamics of the combustion zone and are applicable in the low frequency regime should apply to non-acoustic as well as acoustic instability. Several recent papers (Ref. 18-20) have discussed NAI in terms of the response function normally associated with acoustic instability which evolved from the above models. In the previous report (Ref. 1) the analysis relating to the combustor was separated from the analysis of the combustion so that theoretical combustion models could be combined with the combustor analysis quite readily. A method was also presented for reducing NAI data (obtained from L^* burners) in terms of the response function and the phase of the combustion. In the present report, some of the same NAI data are compared with response function curves calculated from models which are directly applicable to the low frequency regime but which have normally been applied to acoustic instability. For this purpose the model of Denison and Baum (Ref. 16) and a recent model proposed by Hart, Farrell and Cantrell (Ref. 17) have been chosen as being representative and have been utilized to compare the relative applicability of these types of models.

Experimental experience plus the understanding obtained from studies such as outlined above have advanced the level of understanding of NAI so that predictions can be made as to the general behavior of a given propellant. However, there are many instances where the behavior has been different than expected. For example, the one-dimensional models predict that for a given propellant and a given set of conditions, pressure oscillations should occur at one particular frequency. However, two previous instances are known (Ref. 1 and 21) where propellants have oscillated at two different frequencies, under essentially the same conditions, and in each case one of the frequencies was much different than expected. A close scrutiny of the propellant characteristics and the conditions

under which the anomalies occurred has led to the belief that the oxidizer particle size was a major factor producing the unexpected frequency. This provided the motivation which led to the study which is outlined in Section 4.5.

4.2. NONACOUSTIC COMBUSTOR ANALYSIS

In NAI it appears that the combustion couples with the flow out of the rocket chamber in order to induce and sustain instability. Thus, the mass balance for a rocket motor containing burning propellant and exhausting through a sonic nozzle can be written

$$\rho_r A_b - \frac{p A_t}{c^*} = \frac{d}{dt} (\rho_g V_c) \quad (4.1)$$

where c^* is the characteristic velocity for the products of combustion. Assuming ideal gas conditions, a thin combustion zone, a spacewise isothermal gas, and the density of the propellant to be much greater than that of the gas, Eq. 4.1 can be written (For a more complete derivation of Eq. 4.1 and 4.2, see Ref. 18 and 22)

$$\frac{\dot{r}}{\bar{r}} - \frac{\dot{p}}{\bar{p}} = \tau_{ch} \frac{d(p/\bar{p})}{dt} \quad (4.2)$$

Applying standard perturbation techniques and considering an exponentially growing, sinusoidal perturbation such as

$$r = \bar{r} \left(1 + \left(\frac{r'}{\bar{r}} \right) e^{i(\omega - i\alpha)t} \right) \quad (4.3)$$

Equation 4.2 reduces to

$$\frac{r'/\bar{r}}{p'/\bar{p}} = 1 + \alpha \tau_{ch} + i \omega \tau_{ch} \quad (4.4)$$

where α is a growth coefficient and τ_{ch} represents the residence time of the chamber and is directly proportional to L^* . The left hand side of Eq. 4.4 is the response of the mass flow rate in the combustor to the pressure of the combustor and is equal both in magnitude and phase to a similarly defined response of the combustion. All of the terms on the right hand side of Eq. 4.4 are quantities that can be determined experimentally in an L^* -type of burner. Thus, the nonacoustic response of the combustor can be determined experimentally and compared with the

response of the combustion as determined from models such as those of Denison and Baum and Hart, Farrell and Cantrell.

Figure 4.1 is a schematic of an L* burner (Ref. 18, 21, and 22) from which nonacoustic instability data can be obtained. Data have been obtained for two propellants and reduced to the format suggested by Eq. 4.4 in order to make a comparison with one-dimensional models. Both are composite propellants but vary widely in compositions and properties. A-35 is a polyurethane-AP propellant, containing 75% AP, while Utah-TF is a PBAA-AP-Al propellant containing 5% Al, 18% PBAA, 2% copper chromite burning rate catalyst and the remainder AP (see Ref. 1 and 21).

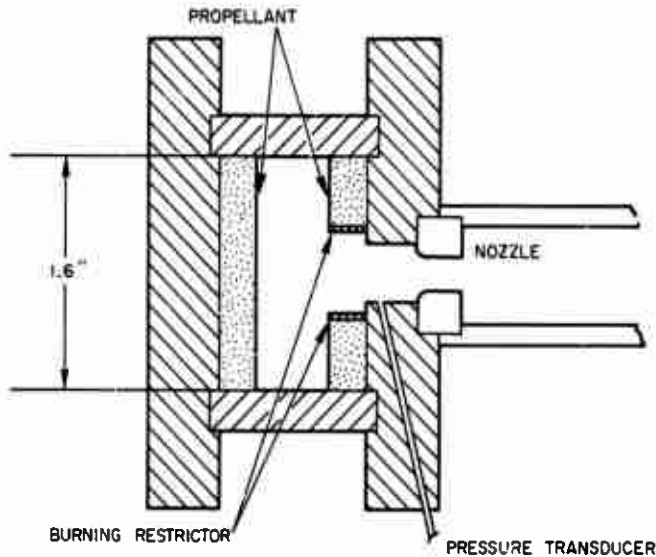


FIG. 4.1. The L* Burner.

4.3. COMBUSTION DYNAMICS MODELS

The model of Denison and Baum (denoted DB) was chosen to provide a basis of comparison for several reasons. First of all it applies specifically to the low frequency range that is of interest in NAI. Secondly, as has been pointed out by Culick (see Appendix A of Ref. 1), the form of the derived response function is extremely simple, containing only two parameters (A and B_{pg}) and the independent variable Ω . Culick has also shown that virtually all of the one-dimensional models reduce to the same function as the DB model. The model of Hart, Farrell, and Cantrell (denoted HFC) was also chosen because it too is directly applicable to

the low frequencies of interest in the present study, and because it contains the effects of a pressure dependent surface reaction.

4.3.1. Comparing the Models

The assumptions involved in both of the analyses are very similar. Both assume a homogeneous, one-dimensional solid, quasi-steady gas phase, and are applicable in the same low frequency range. The principal difference in the models (other than the actual method of analysis) is that Denison and Baum arrive at a burning rate law by starting with the conservation equations and reducing the complexity of these equations with several assumptions. In so doing, the effects of diffusion in the gas phase are included (however, the Lewis number is assumed to be unity). In the original Hart-McClure model (Ref. 14), and in the HFC model, the burning rate is assumed to be a function only of the pressure and the temperature gradient in the gas phase and the latter is then related to the initial temperature of the propellant. By so doing, diffusion in the gas phase is circumvented. The HFC model also takes into account the effects of a surface reaction between a gas phase component and the solid surface. However, the resultant response function can easily be reduced to what will be referred to here as the thermal case (essentially the results derived by Bird, et al (Ref. 15) and in the same form as the Denison-Baum results), or in other words, the analysis without the pressure dependent surface reaction. Throughout the remainder of this section the HFC model will be considered as containing two separate cases, one with and one without the surface reaction.

If the response functions of both models (initially considering the thermal case for the HFC model) are reduced to a common set of nomenclature and put in the form suggested by Culick (see Appendix A of Ref. 1), it is found that there is very little difference. The DB response function is

$$\frac{1}{n} \frac{r'/\bar{r}}{p'/\bar{p}} = \frac{AB_{DB}}{AB_{DB} + \frac{A}{\lambda} + \lambda - (A + 1)} \quad (4.5)$$

and the HFC response function is

$$\frac{1}{n} \frac{r'/\bar{r}}{p'/\bar{p}} = \frac{A'B_{HFC} + \frac{\lambda-1}{n}}{A'B_{hfc} + \frac{A'}{\lambda} + \lambda - (A' + 1)} \quad (4.6)$$

where λ is the characteristic root of the energy equation for the solid and is equal to

$$\lambda = 1/2 [1 + (1 + 4i)^{1/2}] \quad (4.7)$$

It is apparent that the two response functions are essentially the same except for a term in the numerator. This term results from the fact that the HFC model includes pressure dependent surface reactions. If the thermal case of the HFC model is considered (i.e., neglecting the surface reaction) the response function reduces to

$$\frac{1}{n} \frac{r'/\bar{r}}{p'/\bar{p}} = \frac{AB_{HFC}}{AB_{HFC} + \frac{A}{\lambda} + \lambda - (A + 1)} \quad (4.8)$$

Equations 4.5 and 4.8 are identical, differing only in the definition of B. For the two models the definition of this parameter is

$$B_{DB} \equiv \frac{C_p T_f}{C_s (\bar{T}_s - T_o) \left(n + 1 + \frac{E_f}{2RT_f} \right)} \quad (4.9)$$

and

$$B_{HFC} \equiv \frac{T_o}{j(\bar{T}_s - T_o)} \quad (4.10)$$

The differences in the two definitions can readily be traced to the basic difference in the two models involving the form of the burning rate law. In the DB model (as has been mentioned) a burning rate law is derived from the energy and species conservation laws which relate the burning rate to the pressure and the flame parameters. In perturbation form the term

$$\frac{n + 1 + E_f/2RT_f}{T_f}$$

is introduced. It has also been pointed out that in the HFC model the burning rate is assumed to be a function of the pressure and the temperature gradient in the gas phase. The burning rate is then written as a perfect differential in terms of the appropriate pressure and gradient terms. The latter is then related to the initial temperature of the propellant which results in the factor j/T_o .

$$\left(\text{where } j \equiv \frac{T_o}{T} \frac{\partial \bar{r}}{\partial T_o}\right).$$

Thus, the only real difference in the two models being compared (considering the HFC thermal case only) is in the burning rate law that is employed, and for the sake of numerical calculations, the models are identical. The only difference lies in relating the original parameters from the respective models to the value of B.

4.3.2. Comparing Calculated Response Functions and Data

The purpose of this section is to compare the predictions of the chosen instability models with the results obtained from actual NAI data. In order to evaluate Eq. 4.5, 4.6, and 4.8, values for A and B must be obtained. Values of the individual parameters have been measured or obtained from the literature wherever possible, and most probable ranges of values have been obtained where a precise value is not available. These are tabulated in Table 4.1, with literature references given, where available. The corresponding parameter values are also tabulated in Table 4.1 where possible.

TABLE 4.1. Values of Parameters for Numerical Calculation

	Representative values	Values used in calculations	Corresponding parameter values
T_o	300°K	300°K	...
n	0.46 ^a and 0.49 ^b	0.50	...
T_a	750 to 1,000°K ^{23,24} ^c	900°K	...
E_a	22 ²⁵ and 30 ²⁶ Kcal	22, 30 Kcal	A = 8, 11
j	0.6 to 0.7 ^{27,28}	0.65	B _{HFC} = 0.77
T_f	2160 ^d and 2860°K ^e	2160, 2860°K	See Fig. 4.2
E_f	10 to 50 Kcal	10, 30, 50 Kcal	See Fig. 4.2

^a Measured for A-35 propellant.

^b Measured for UTAH-TF propellant (See Ref. 22).

^c Superscripta refer to references.

^d Calculated adiabatic flame temperature for A-35 propellant.

^e Calculated adiabatic flame temperature for UTAH-TF propellant.

Figures 4.2, 4.3, and 4.4 show how the nondimensional parameters A and B vary as functions of the physical parameters. In determining B_{DB} it was assumed that the specific heat of the gas phase is equal to that of the solid, and that the mean flame temperature, the mean surface temperature and the burning rate exponent, n , are all constant. Substituting the values of Table 4.1 into Eq. 4.9, the range of B_{DB} is from 0.55 to 2.0 for the two propellants. This range of values reflects the uncertainty in the value of the activation energy of the flame which is the parameter having the greatest uncertainty. This is graphically demonstrated in Fig. 4.2.

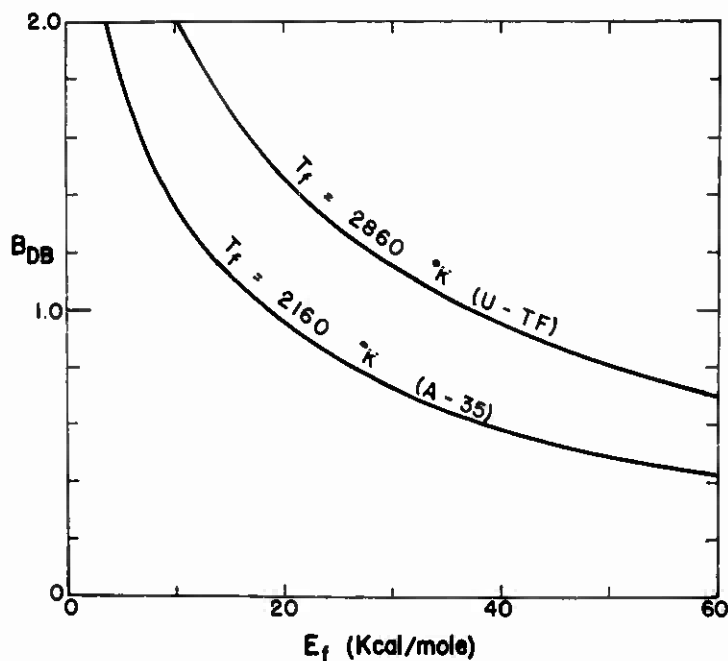


FIG. 4.2. The Denison and Baum Gas Phase Parameter B as a Function of E_f With the Flame Temperature as a Parameter.

From Fig. 4.3, it is seen that the parameter B_{HFC} does not vary greatly but is close to 0.8 for reasonable values of the parameters. The values of the parameters in Table 4.1 result in $B_{HFC} = 0.77$ which was subsequently used in all the calculations for that model. In Fig. 4.4 A would appear to have a value on the order of 10. Combining the

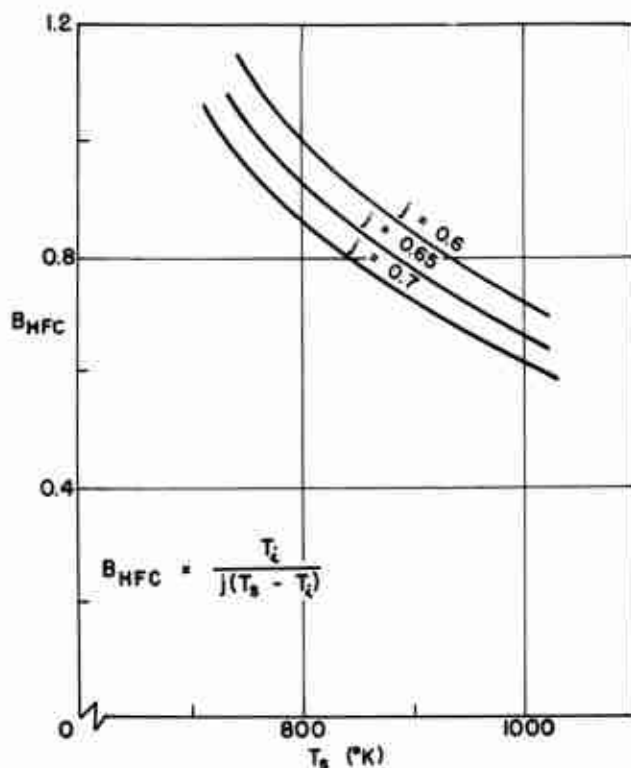


FIG. 4.3. The Hart, Farrell, Cantrell Parameter B as a Function of T_s With the Temperature Sensitivity j as a Parameter.^s

values of the various parameters given in Table 4.1 results in values of approximately 8 and 11 for the nondimensional parameter Λ , and these values (for a constant surface temperature of 900°K) have been used in the calculations for both models.

Using the values of the various parameters as noted above, the real part and the phase of the response function have been calculated from Eq. 4.5, 4.6, and 4.8 and are compared with the data for one of the particular propellants in each of the following figures. Thus, Fig. 4.5 and 4.6 contain the data of A-35 propellant (Ref. 1) and the curves calculated from the models using the parameters that are indicated in the

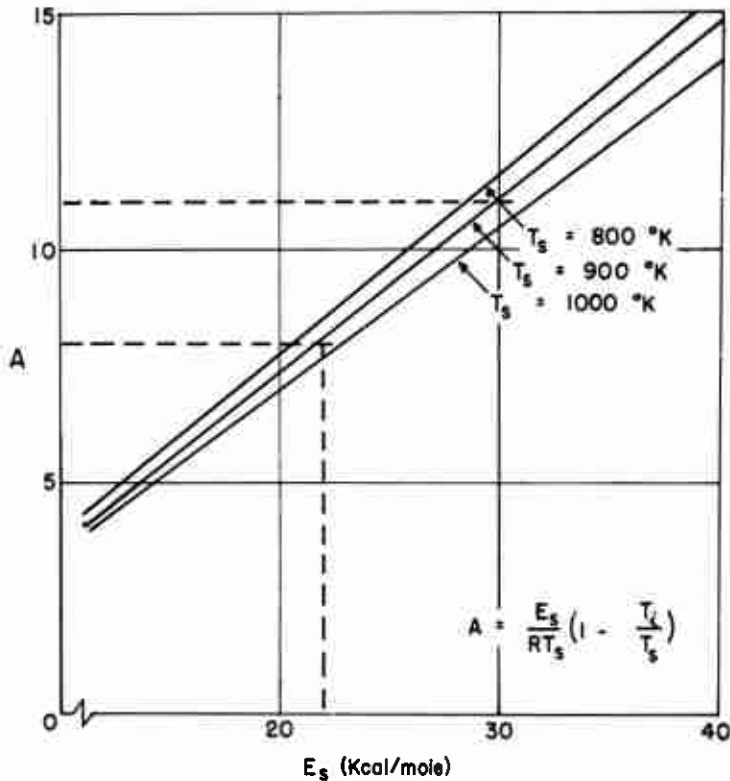


FIG. 4.4. The Solid Phase Parameter A Plotted Versus E_s With the Surface Temperature as a Parameter.

figures. It should be noted here that even though the scatter in the data is considerable, the fact that a comparison can actually be made between data of this nature and the theoretical models is considered significant. In comparing the two different cases for the HFC model, it is apparent that the addition of a pressure-dependent surface reaction greatly enhances the predicted response of the propellant.

Of the calculations for the DB model, only the curve corresponding to the flame-activation energy of 30 Kcal/mole is included in the two plots. The curves for $E_f = 10$ Kcal/mole resulted in responses that were entirely too low to predict instability, while the curves for $E_f = 50$ predicted exponential instability (nonsinusoidal instability; see the discussion in Ref. 16 on this subject). Therefore, these curves were not included in Fig. 4.5 and 4.6. Although each of the curves is quite

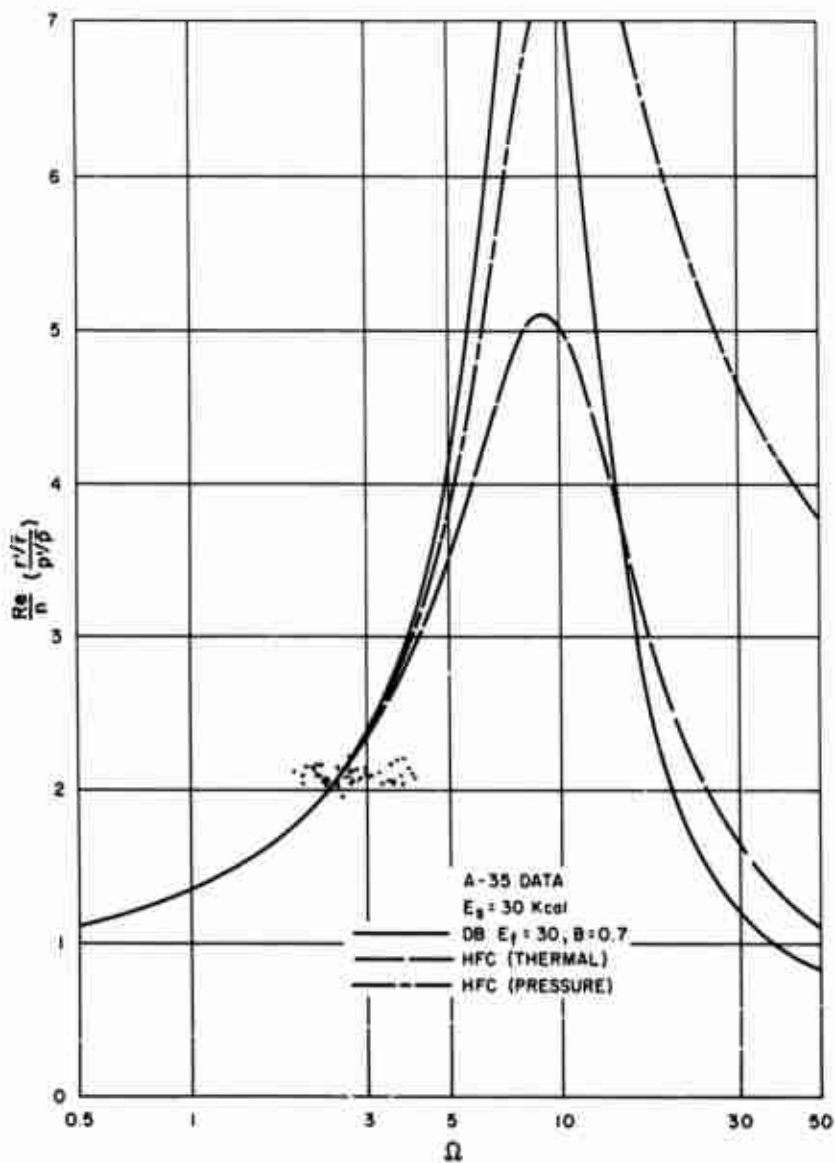


FIG. 4.5. Response Functions as a Function of the Nondimensional Frequency, Ω , and for the Given Parameters Compared With A-35 Data. HFC (thermal) refers to the HFC model, thermal case. HFC (pressure) refers to the HFC model including the pressure dependent surface reaction term.

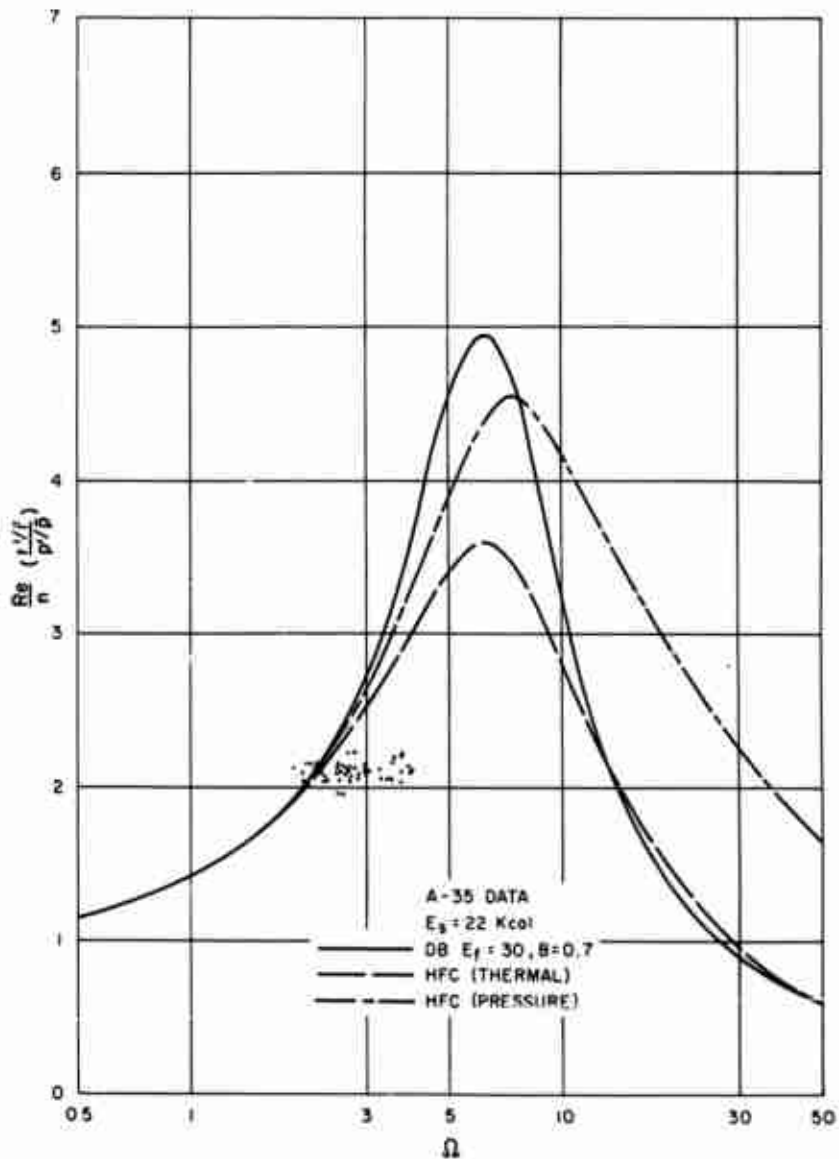


FIG. 4.6. The Response Functions for $E_s = 22$ ($A = 8$) Compared With A-35 Data.

different from the others it would be impossible to judge from the data which set of parameters more nearly predicts actual behavior. However, the phase has also been calculated for these parameters and is compared with the L* burner data in Fig. 4.7 and 4.8. Again, the scatter is considerable, but a much more definite trend is apparent. On the basis of these latter figures, it can be concluded that for both models the curves corresponding to $E_s = 22$ Kcal/mole and, in particular, the purely thermal part of the HFC model, provide the better fit with the data. The results from these latter two curves will be used later in a comparison with basic NAI data.

Figures 4.9 through 4.12 contain the Utah TF propellant data and are compared with the theoretical curves. Two curves for the DB model have been included, corresponding to $E_f = 30$ and 50. In Fig. 4.9 and 4.10 the value of 50 provides the better fit with the data. The curves for $E_f = 30$ are both much lower than the data. Based on these two figures, it would appear that the surface-activation energy of 50 in both models provides the better fit of the data. Turning again to the calculated values of the phase, Fig. 4.11 and 4.12 provide additional information upon which a judgement may be based. In Fig. 4.11, the curve that most closely fits the data corresponds to the DB model with $E_f = 30$, but this set of parameters has already been rejected on the basis of Fig. 4.8. From Fig. 4.12 it is again concluded that the best fit of the data is obtained for $E_s = 22$ for both models, utilizing the thermal part of the HFC model ($B = 0.77$) and $E_f = 50$ for the DB model ($B = 0.8$).

In summarizing the results of this section, it seems appropriate to choose the sets of parameters for the calculations that result in the best fit of the data for the two models to be coupled with NAI combustor theory. The only parameter that was varied in the HFC model was the surface-activation energy and the results indicate that the value of 22 Kcal/mole ($A = 8$) gives the best results for both propellants. It also appears that the thermal case of the HFC model is more applicable than the pressure-dependent case. This observation is not apparent from an examination of the response function results alone as the real part of the two cases does not vary much at low frequencies. However, the phase is different over the range of frequencies and upon this basis, it is possible to draw the conclusion that the thermal case is more applicable. With the DB model the surface-activation energy of 22 Kcal/mole also gave the best results for both propellants. This would be anticipated because the surface-activation energy is contained in the parameter A in exactly the same manner for both models. However, for the TP propellant a flame-activation energy of 50 Kcal/mole gave the best fit while the value of 30 Kcal/mole was best for A-35. A mechanistic argument explaining the difference between the two propellants is beyond the scope of this report and will not be attempted at this time. This difference in flame-activation energy is not so dramatic when the pronounced differences of the propellant compositions are taken into account.

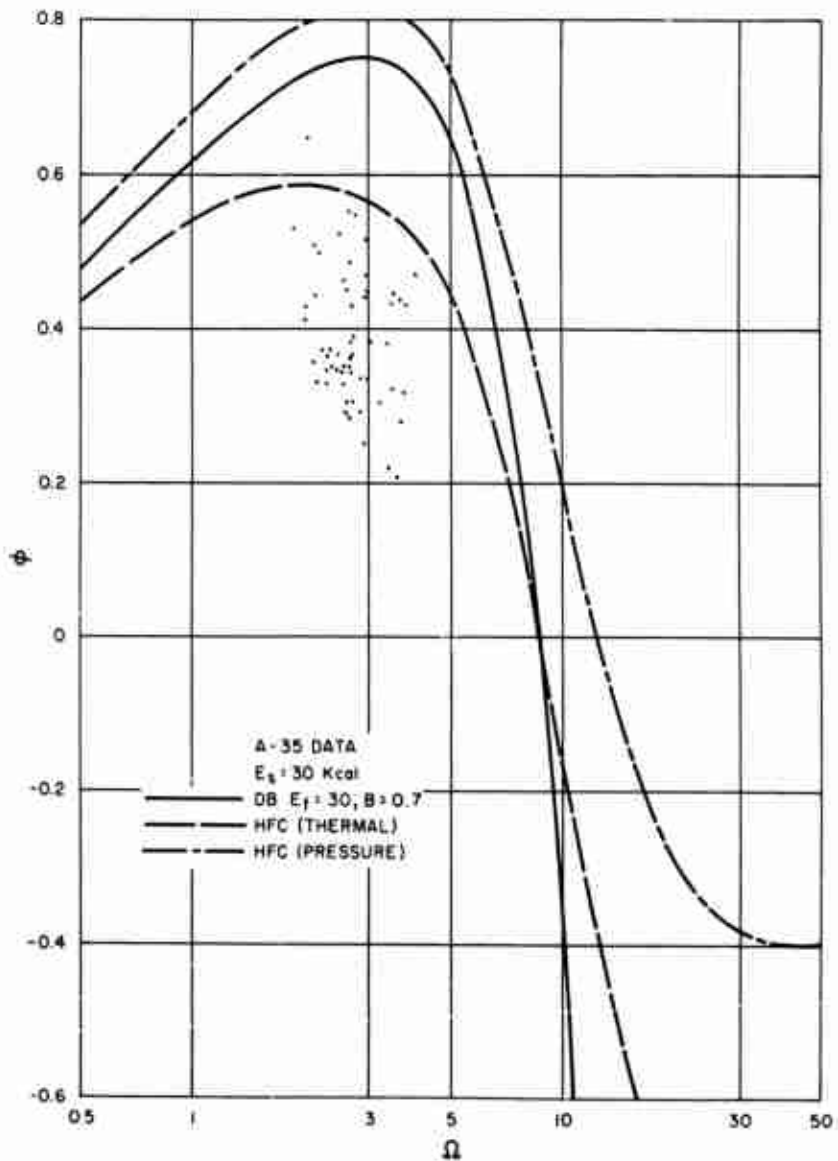


FIG. 4.7. Theoretical Phase Versus Nondimensional Frequency for $E_s = 30$, ($A = 11$), Compared to A-35 Data.

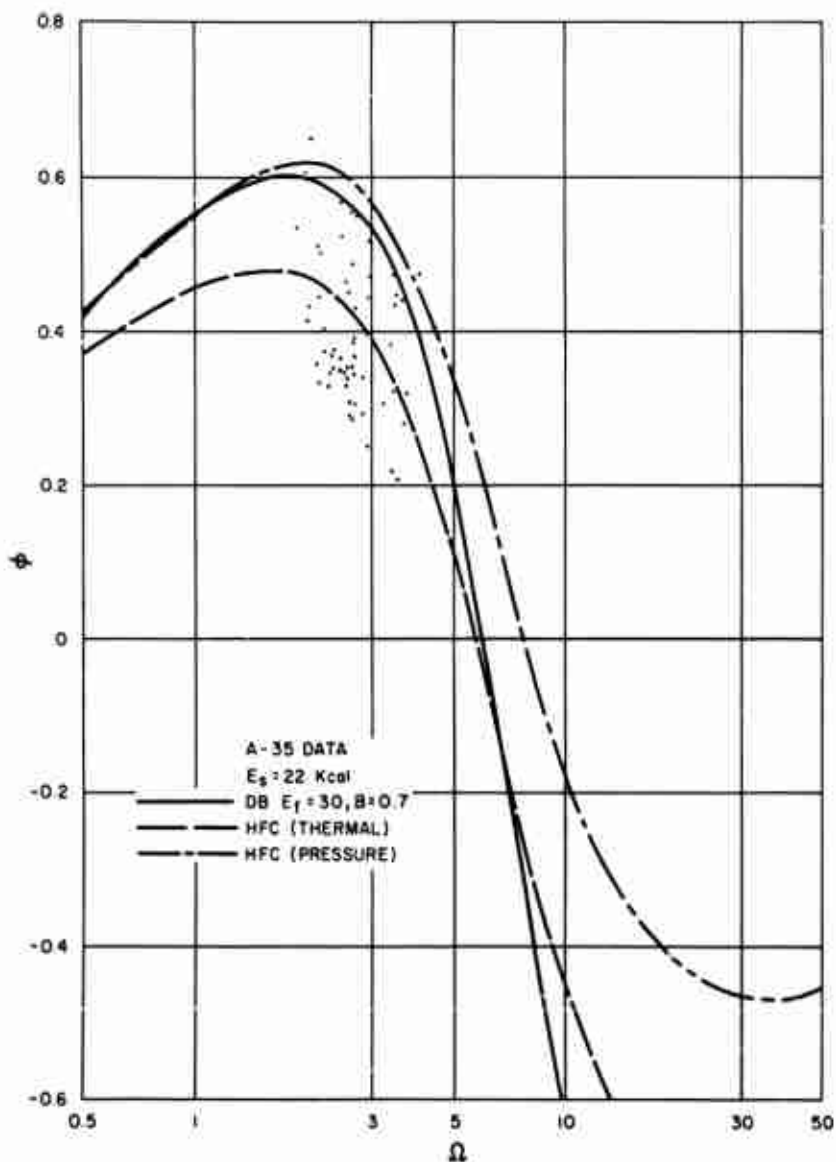


FIG. 4.8. Theoretical Phase Versus Ω for $E_s = 22$, ($A = 8$), Compared to A-35 Data.

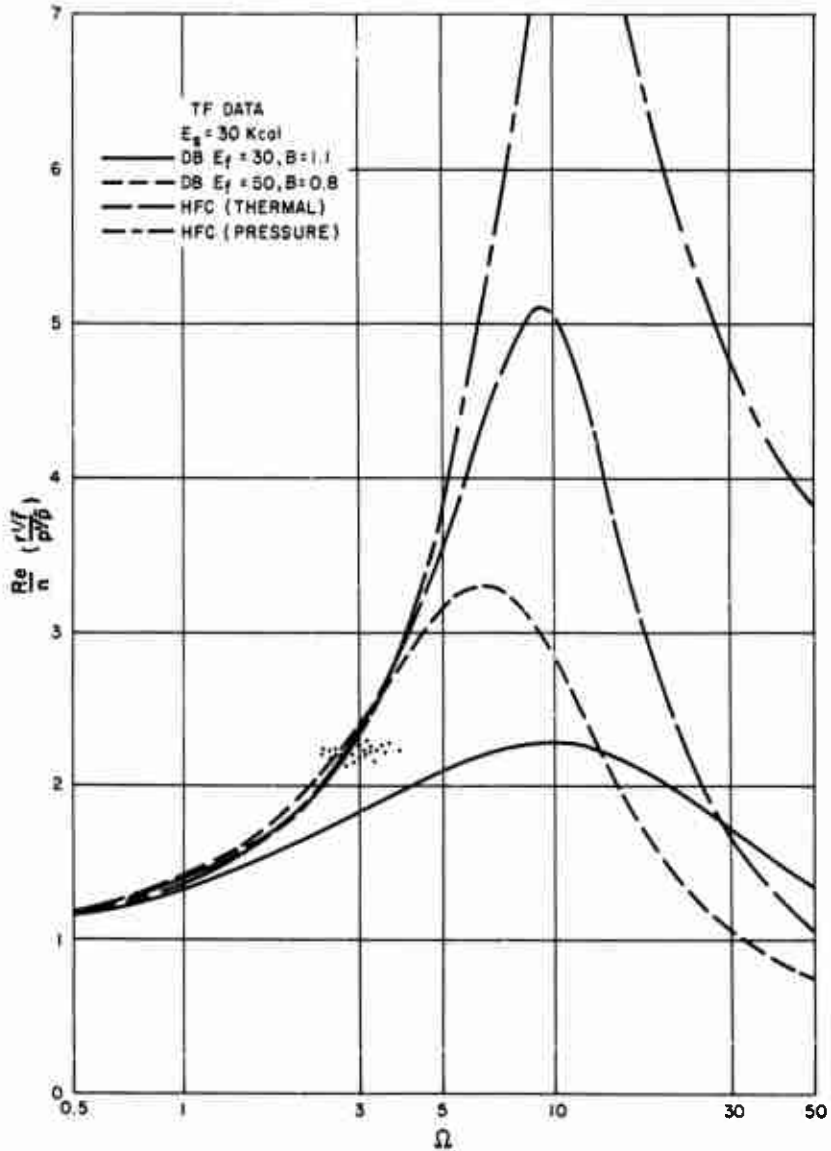


FIG. 4.9. The Response Functions for $E_s = 30$, ($A = 11$), Compared With Utah-TF Data.

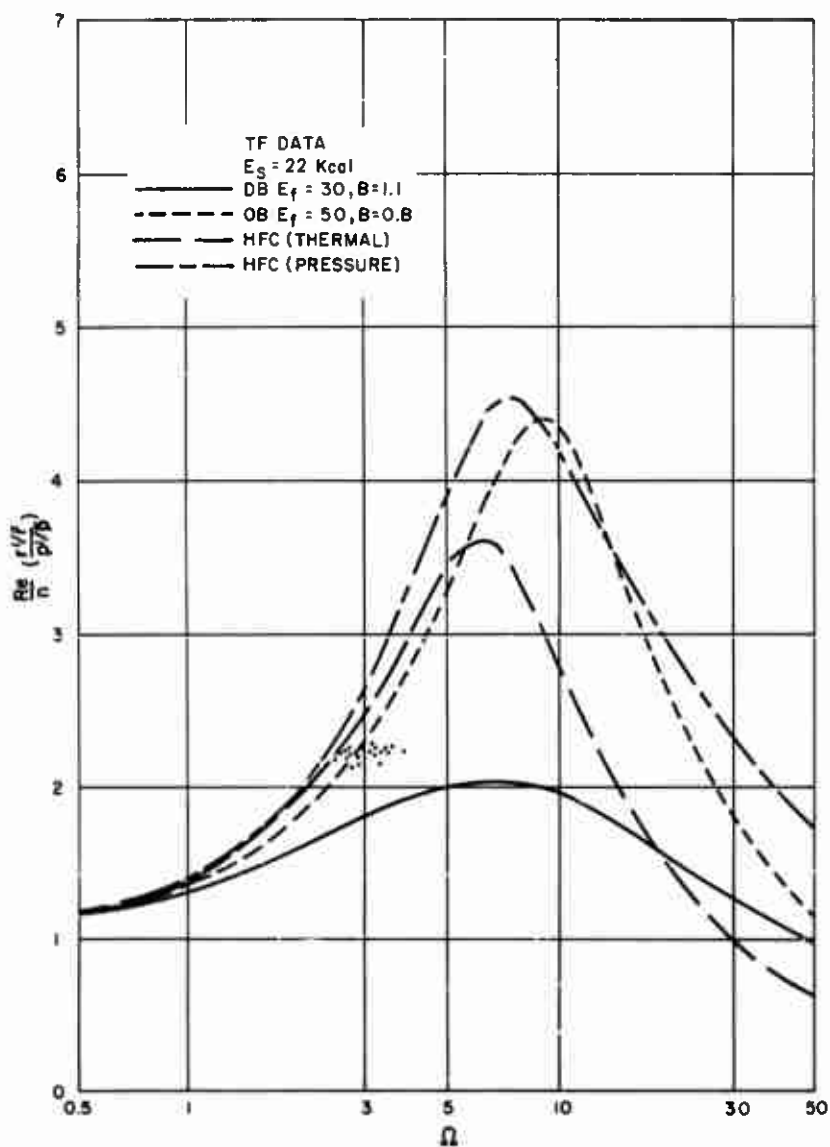


FIG. 4.10. The Response Functions for $E_s = 22$, ($A = 8$), Compared With Utah-TF Dats.

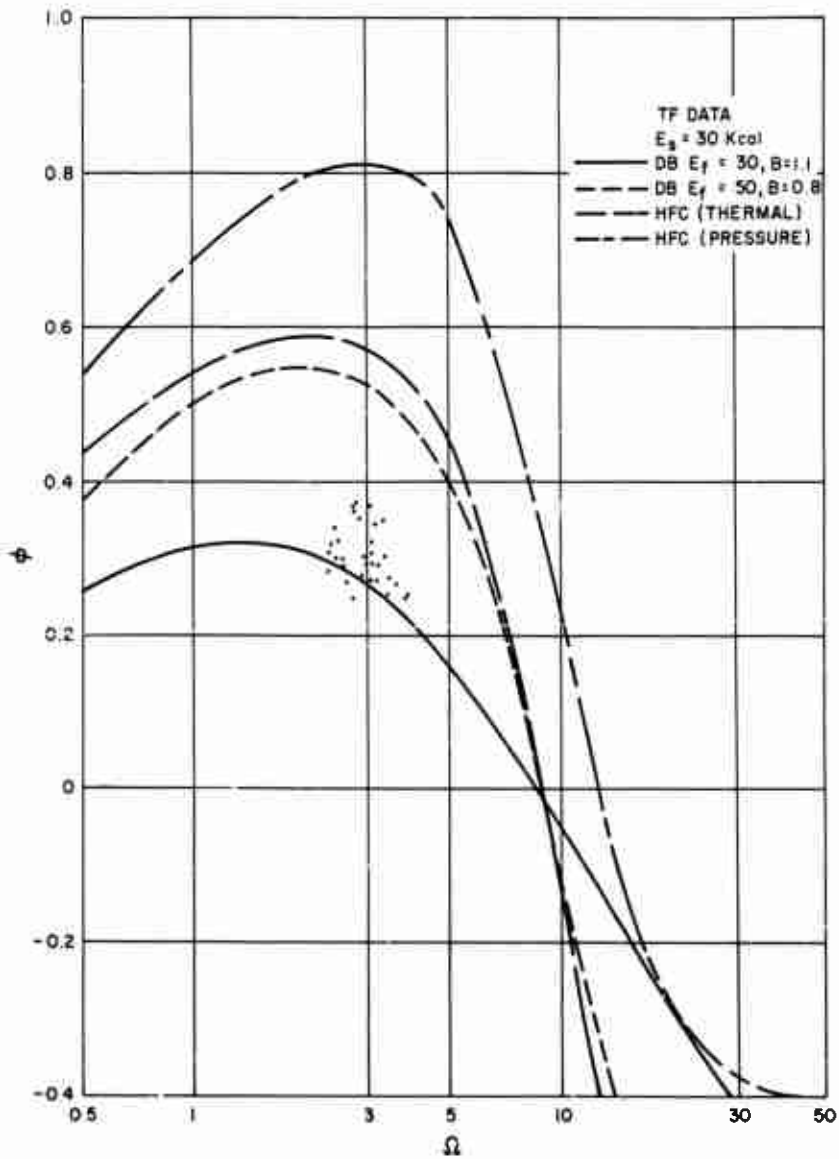


FIG. 4.11. phase Versus Ω for $E_s = 30$, ($A = 11$), Compared to Utah-TF Data.

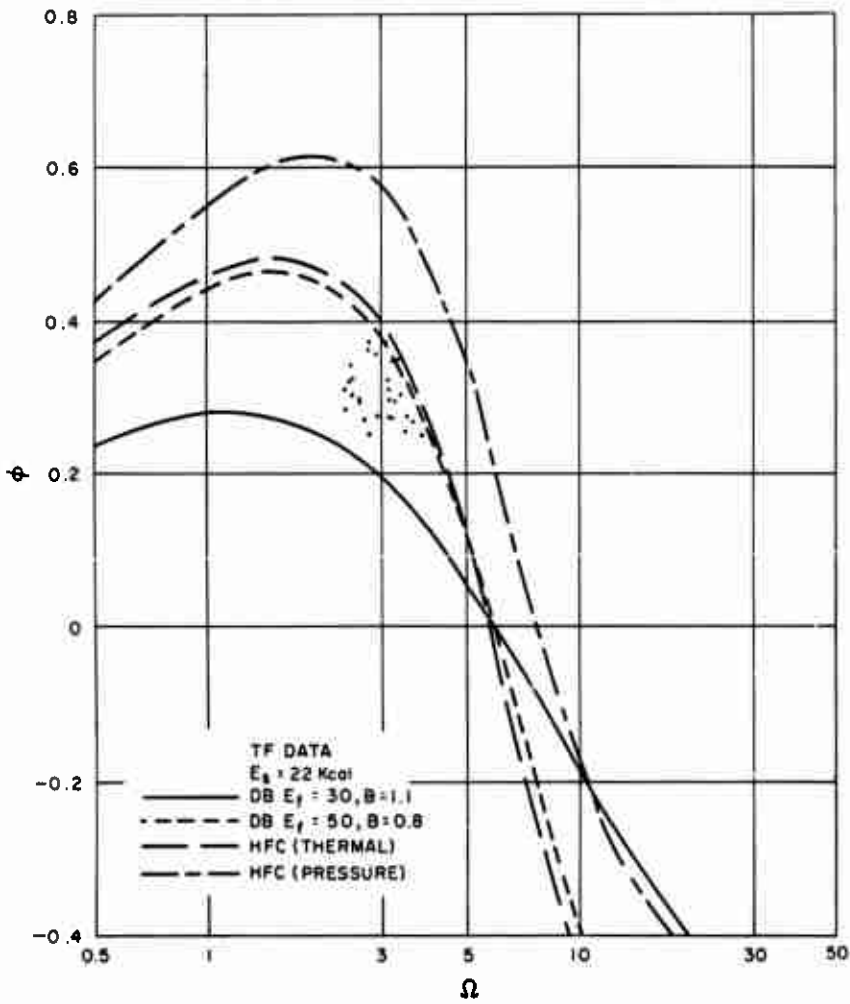


FIG. 4.12. Phase Versus Ω for $E_s = 22$, ($A = 8$), Compared to Utsh-TF Data.

It should be noted here that the value of B in these two cases is 0.8 and 0.7, respectively, while B_{HFC} was 0.77. Due to the numerical identity of the models there is essentially one set of nondimensional parameters that give the best fit for both propellants and for both models, i.e., $A = 8$, $B = 0.7-0.8$.

4.4. NAI CHARACTERISTICS FROM THEORETICAL MODELS

If the various parameters discussed in the previous section were known a priori, the response function could be calculated directly from the models. This information could then be combined with Eq. 4.2 to give the nonacoustic instability characteristics of a rocket motor or a laboratory burner.

In the following, the NAI characteristics for the two propellants studied will be calculated utilizing the models and the apparently best parameters as determined from Fig. 4.5 through 4.12. This will permit the direct comparison of the one-dimensional models with experimental data from an L^* burner. Because of the numerical similarity between the two models only one set of parameters was used in the calculations for each propellant (for A-35: $A = 8$, $B = 0.77$ and for Utah TF: $A = 8$, $B = 0.8$). From Eq. 4.4

$$\operatorname{Re} \left(\frac{\bar{r}'}{\bar{p}'} \right) = 1 + \alpha \tau_{ch} \quad (4.11)$$

and

$$\tan \phi = \frac{\omega \tau_{ch}}{1 + \alpha \tau_{ch}} \quad (4.12)$$

where ϕ represents the phase of the combustion relative to pressure. Combining these equations and nondimensionalizing them with the thermal wave time constant after solving for τ_{ch} yields

$$\frac{\tau_{ch}}{\tau_{tw}} = \frac{\operatorname{Re} \left(\frac{\bar{r}'}{\bar{p}'} \right) \tan \phi}{\omega \tau_{tw}} = \frac{\operatorname{Re} \left(\frac{\bar{r}'}{\bar{p}'} \right) \tan \phi}{\Omega} \quad (4.13)$$

The right-hand side of the equation can be completely determined theoretically from the models, thus relating the nondimensionalized L^* ($\tau_{ch} \propto L^*$) to the nondimensional frequency. Similar relationships have been derived previously (Ref. 22), but without considering the details of the gas-phase combustion. Utilizing the parameters for the given propellants as discussed above, values for the response function and phase as a function of the nondimensional frequency can be substituted

into Eq. 4.13 to give the nondimensional, L^* -frequency relationship. In Fig. 4.13 theoretical curves of this nature are compared with the actual data for the two propellants. The theoretical curves are much more curved than one would expect from the trend of the actual data, but other than this, the agreement is quite reasonable.

Because of the intimate relationship between the growth constant and the stability of the system, it would be worthwhile to have a theoretical relationship describing the growth constant. This can be determined readily by solving for α from Eq. 4.12 obtaining

$$\alpha = \frac{\omega}{\tan \phi} - \frac{1}{\tau_{ch}} \quad (4.14)$$

Evaluating τ_{ch} from Eq. 4.13 and dividing through by the frequency yields

$$\frac{\alpha}{\omega} = \frac{\operatorname{Re}\left(\frac{r'/\bar{r}}{p'/\bar{p}}\right) - 1}{\operatorname{Re}\left(\frac{r'/\bar{r}}{p'/\bar{p}}\right) \tan \phi} \quad (4.15)$$

Again, all of the terms on the right-hand side of the equation can be determined from the theoretical models. Using the parameters noted above, results obtained from Eq. 4.15 are compared with actual data in Fig. 4.14. Within the limits of the data scatter, the theoretical prediction is excellent.

If the growth constant is now set equal to zero, the stability limit for NAI is obtained. From Eq. 4.4 and 4.15 this condition is seen to be

$$\left[\operatorname{Re}\left(\frac{r'/\bar{r}}{p'/\bar{p}}\right) = 1\right]_{\text{stability limit}} \quad (4.16)$$

From Fig. 4.14 and the equations for the response function it is apparent that Eq. 4.16 will hold true for a particular value of the frequency (i.e., Ω_{cr}) for any given set of parameters. Substituting these results into Eq. 4.13 yields

$$\tau_{ch} = \left(\frac{\tan \phi}{\Omega_{cr}}\right) \tau_{tw} \quad (4.17)$$

Thus, it should be possible to represent the stability limit as a straight line passing through the origin on a plot of τ_{ch} versus τ_{tw} .

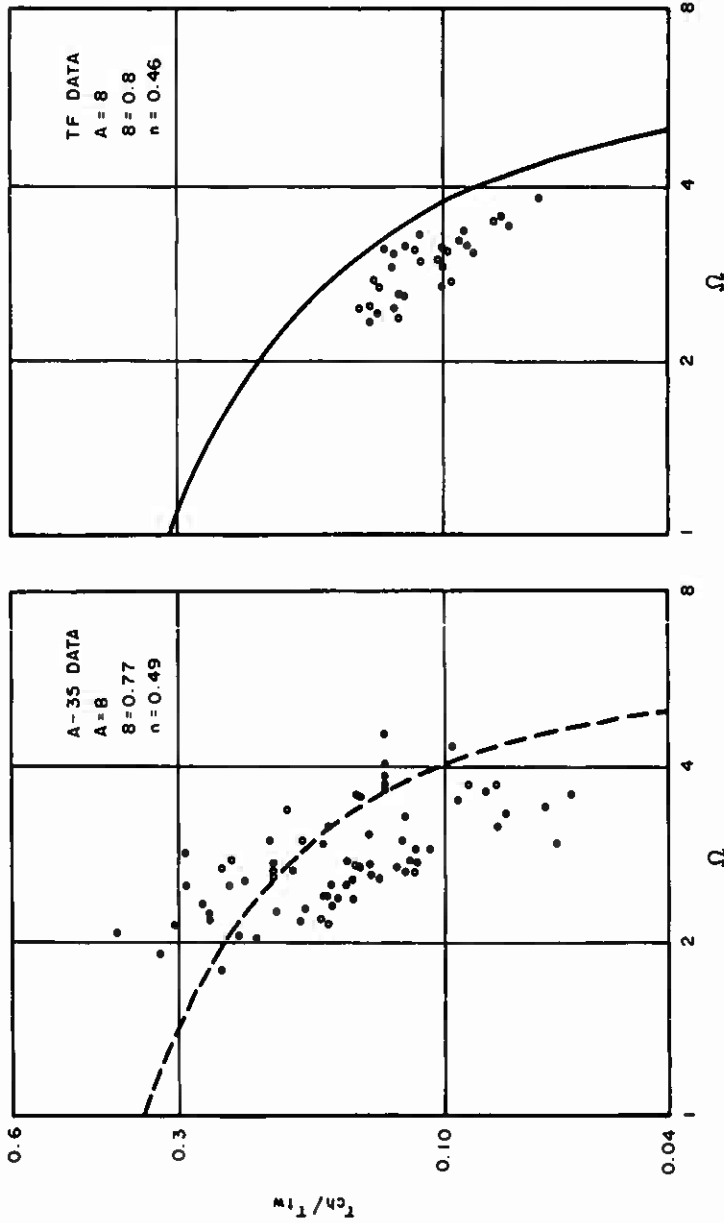


FIG. 4.13. Comparison of Theory (Using the Best Parameters as Determined by the Response Function and Phase Curves) With Experiment. Nondimensional L^* versus nondimensional frequency.

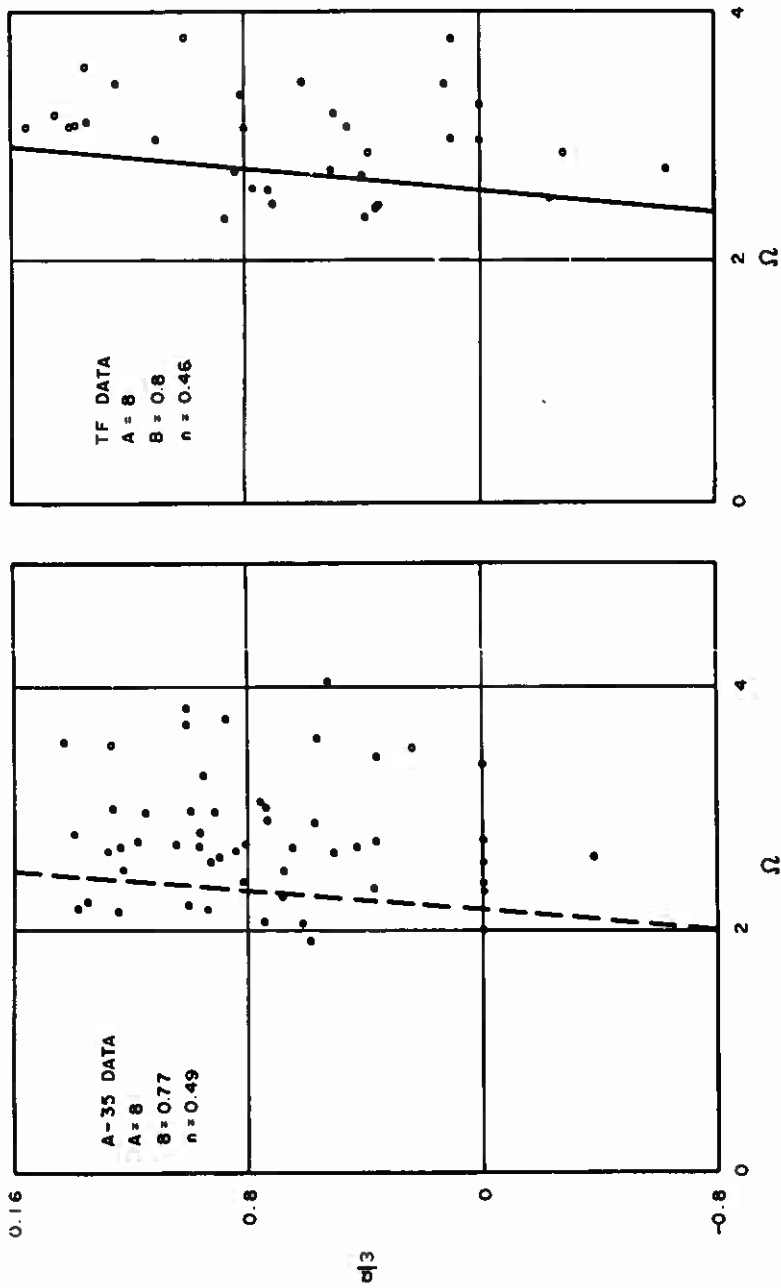


FIG. 4.14. Comparison of Theory With Experiment. Nondimensional growth constant versus nondimensional frequency.

Figure 4.15 contains such a plot--again comparing the results of the models with the actual data. The solid circles represent firings with $\alpha < 0$ (i.e., in a stable regime where developed oscillations decayed). Considering that data can only be obtained in the unstable region and up to the stability limit, a line drawn along the left-hand edge of the data should represent the stability limit. For the Utah-TF propellant, the theoretical results agree quite well with the data. However, for the case of the A-35 data, it would be virtually impossible to draw such a line and have it pass through the origin. Apparently something occurs with this propellant which is not accounted for by the models.

4.5. THE EFFECT OF OXIDIZER PARTICLE SIZE ON NAI

In order to make the models mathematically tractable certain assumptions such as a homogeneous solid burning in one dimension with a quasi-steady gas phase were made. In an effort to assess the validity of the assumption of one-dimensional burning of a homogeneous solid, a program was initiated to study the effects of oxidizer particle size distribution and binder type on the nonacoustic combustion instability. The following sections present the effect oxidizer particle size has on NAI.

4.5.1. The Layer Frequency Concept

An idealized propellant can be visualized as consisting of oxidizer particles stacked one on top of another, surrounded by binder. The characteristic time to burn through one particle is the particle diameter divided by the burning rate (i.e., $\tau = D/r$). If the particles are indeed stacked in layers then this characteristic time will be repeated at a frequency of one cycle per particle, or $f = 1/\tau = r/D$ (Fig. 4.16). If the entire surface of a burning propellant regressed so that each oxidizer particle was exposed at the same time and burned "in phase" with enough other oxidizer particles, then this layer frequency would become a measurable phenomenon, contributing to combustion instability. This phase correlation³ concept is schematically pictured in Fig. 4.17.

In an actual propellant the oxidizer particles are randomly distributed and therefore the phase correlation of the entire surface would be somewhat difficult to achieve and would most likely result in a frequency less than noted above. That is to say,

$$f = K/\tau = Kr/D \quad (4.18)$$

³ This "phase correlation" concept was first proposed by Price (Ref. 29) in connection with the shedding of agglomerated metal from a propellant surface.

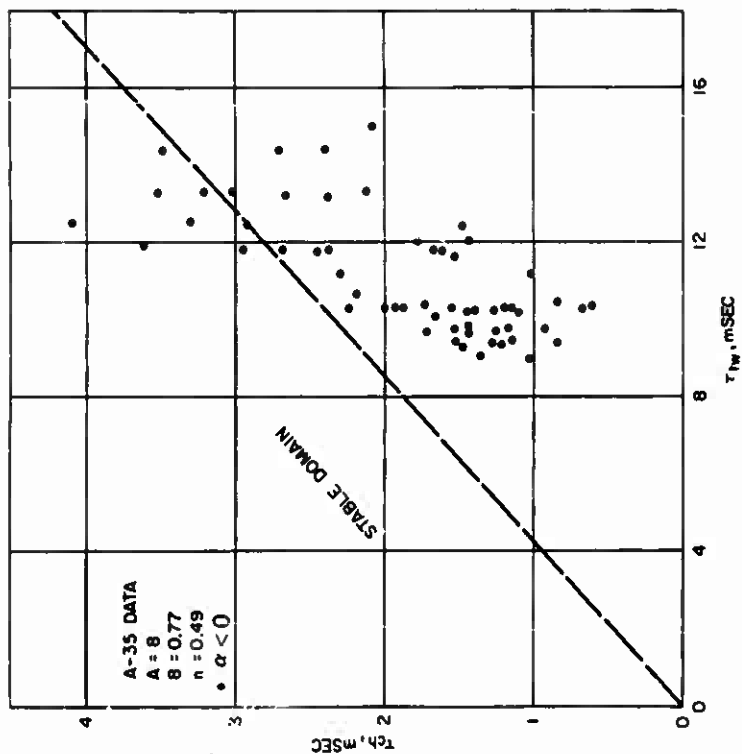
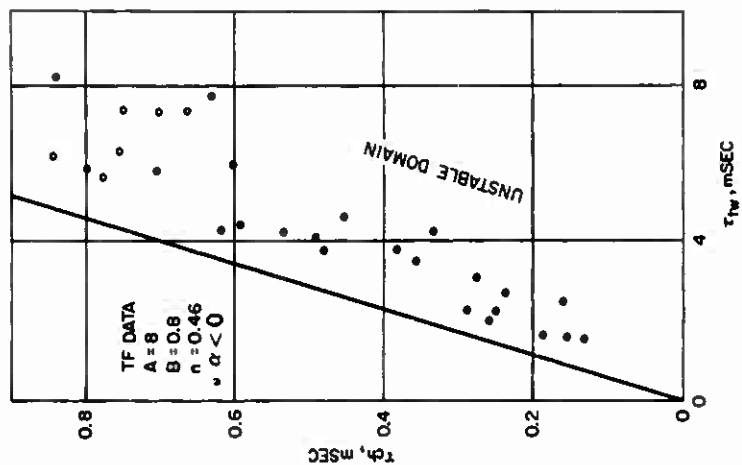
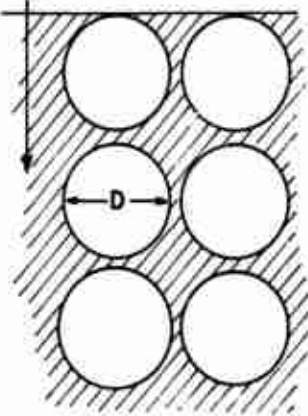


FIG. 4.15. Comparison of Theories With Experiment. Stability limit-chamber residence time versus thermal wave relaxation time. To the right of the lines is the unstable domain.

Direction of Burn



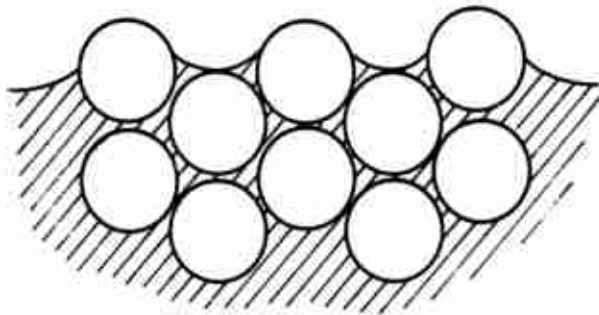
characteristic time = $\frac{\text{oxidizer particle size}}{\text{burning rate}}$

$$\tau = \frac{D}{r} \text{ (sec)}$$

frequency = $\frac{(\text{constant})(\text{one cycle/particle})}{\text{characteristic time/particle}}$

$$f = \frac{K}{\tau} = \frac{Kr}{D}$$

FIG. 4.16. The Layer-Frequency Concept--The Frequency at Which a Combustion Wave Burns Through Layered Particles is Directly Proportional to the Burning Rate and Inversely Proportional to the Diameter.



Phase correlated
surface with
particles burning
in phase

FIG. 4.17. Phase Correlation--If the Burning Surface Arranges Itself Such That Sufficient Oxidizer Particles are Exposed Simultaneously, Then the Surface is Said to be "Phase Correlated", and Measurable Oscillations will Occur at the "Layer Frequency".

where $\tau < 1$ due to the fact that the oxidizer particulae are not attacked uniformly one on top of the other but are randomly oriented. The possibility of having a phase correlated surface can be explored by examining the surface of a quenched propellant. Figure 4.18 is a photomicrograph of the surface of a propellant that extinguished itself while chuffing. The protruding surfaces of the oxidizer particulae are readily apparent and it is not unrealistic to imagine that the surface was indeed phase correlated to some degree. The extent to which a surface must be "correlated" in order for instability to be observable is a question that could be pursued in the future.

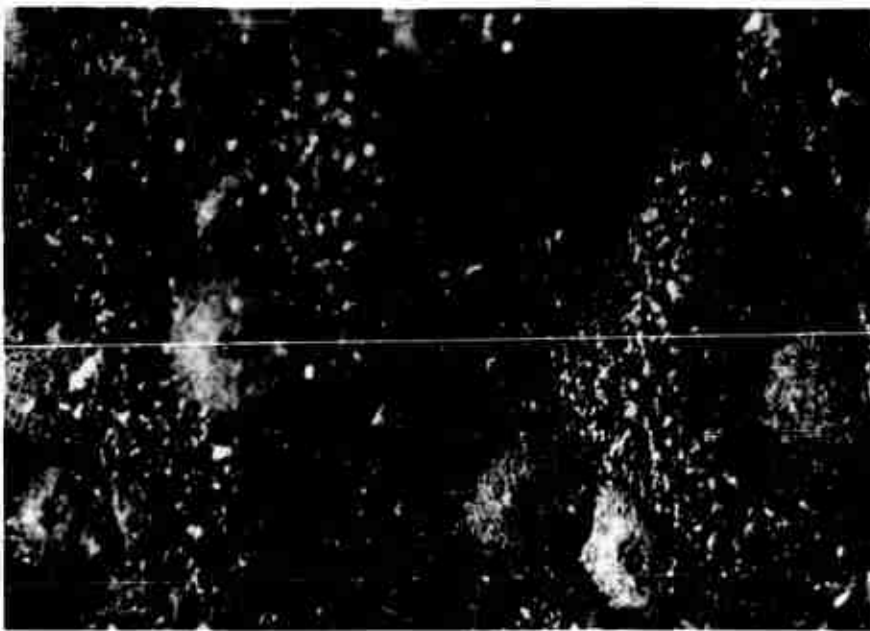


FIG. 4.18. Photomicrograph of an Extinguished Sample of A-155 Propellant. The large, protruding crystals had an initial, mean diameter of 400 μ .

It was pointed out in Section 4.1 that a dual frequency nature had been exhibited by two different propellants, and that this duality had been attributed to the oxidizer particle size. According to the layer frequency concept outlined above, any propellant containing a bimodal blend of oxidizer should, under the right conditions, display this dual-frequency behavior. To check this postulate a series of bimodal AP propellants was made having 25% polyurethane binder and 75% solids (1%

carbon black was used to reduce the translucence of the propellant). Part of the AP in the propellants was a coarse oxidizer and part was finer oxidizer, with the oxidizer particle size distribution for each propellant given in Table 4.2. The AP that was used was the special spherical AP produced by the American Potash and Chemical Corporation, and all blends except the 15 μ were screened to give either ± 18 or $\pm 25\%$ of the mean particle diameter. Chemically, the propellants were the same; only the physical nature was changed by varying the oxidizer particle sizes. In the remainder of this section the results obtained with these propellants and their relationship with the layer frequency will be discussed.

TABLE 4.2. Bimodal Propellant Compositions.

Propellant designation	Ingredients and weight percent				
	Ammonium Perchlorate		Binder		Other
A-146	37.5% 15 μ	37.5% 80 μ	25% polyurethane	
A-148	37.0% 15 μ	37.0% 200 μ	25%	do	1% carbon black
A-149	37.0% 90 μ	37.0% 600 μ	25%	do	1% do
A-151	37.0% 45 μ	37.0% 200 μ	25%	do	1% do
A-155	37.0% 45 μ	37.0% 400 μ	25%	do	1% do
A-156	51.8% 15 μ	22.2% 200 μ	25%	do	1% do
A-157	22.2% 15 μ	51.8% 200 μ	25%	do	1% do
A-158	37.2% 90 μ	37.5% 600 μ	25%	do
A-159	36.0% 15 μ	36.0% 200 μ	25%	do	1% carbon black 1% n-butyl ferro- cene
A-160	36.0% 45 μ	36.0% 400 μ	25%	do	1% carbon black 1% n-butyl ferro- cene

4.5.2. The Non-one-dimensional Nature of Propellant

The models available (Section 4.3) to predict the general behavior of a given propellant all assume that the propellant is a homogeneous solid burning in a one-dimensional manner. The degree of validity of these assumptions may be investigated by examining the heterogeneous, non-one-dimensional nature of the propellant with respect to thermal wave penetration and oxidizer particle size. Figure 4.19 is a schematic showing the relative dimensions of typical oxidizer particles and

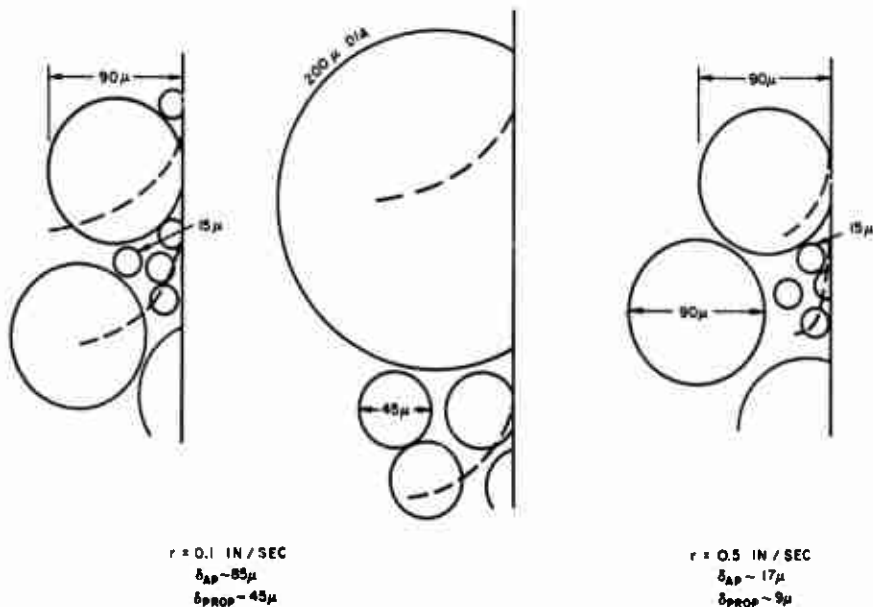


FIG. 4.19. Thermal Waves in Heterogeneous Propellants--The Circles Represent AP Crystals (Drawn to Scale) and the Dashed Lines Represent the Temperature Profile and the Distance that the Profile Penetrates Into the Crystal. The upper profile was calculated from the properties of AP alone and the bottom one is based on the average propellant properties.

temperature profiles for two different burning rates. The thermal profiles which have been included are based on the thermal properties of AP alone (top profile) and on the average properties of the propellant (bottom profile). At low pressure and low burning rate, the thermal wave thickness is quite large, and for small oxidizer particle sizes the propellant can be considered homogeneous and burning may essentially be considered one-dimensional. However, at a burning rate of 0.1 in/sec, the thermal wave thickness is the same order of magnitude as the oxidizer particle sizes used in this study. Hence, the homogeneous and one-dimensional assumptions necessary to the analytical models would appear to be tenuous under these conditions, and at the same time these are the conditions where the layer frequency would most likely be observed. If the thermal wave thickness just matches the particle size, then as the particle burns the combustion wave will soon be passing into preheated AP and the local burning rate of the particle will increase as the

particle burns out. This rapid burning of the remains of an oxidizer particle has been observed in movies such as those described in Section 3. It can readily be imagined that if such a phenomenon could be "phase correlated" over the entire propellant surface, pressure oscillations would be produced.

4.5.3. Results From Bimodal Propellants

From the above discussions the greatest departure from one-dimensional theory would be expected at high values of burning rate (smaller thermal wave) and for large oxidizer particle sizes. This is indeed the case as illustrated by the trends of the data plotted in Fig. 4.20. Test data as well as the burning rates of the various propellants are tabulated in Appendix A. The cross-hatched parabolic band represents the prediction of one-dimensional theory, and the data are for the various propellants. At low values of the burning rate, $r < .09$ in/sec, the data agree moderately well but at higher burning rates ($r > .10$ in/sec), the experimental values deviate quite significantly from the one-dimensional theory.

Whereas Fig. 4.20 is a composite plot, Fig. 4.21-4.26 present the data for each individual propellant. In the figures the shaded areas bounded by straight lines represent the mapping of the equation of $f = r/D$ for the particle size distribution used in the formulation of that propellant. These shaded areas show where the layer frequency should show the strongest effect (for $K = 1$).

Figure 4.21 presents the data for the unimodal, 90 μ AP propellant, A-35. For this propellant the data agree well with both the layer frequency concept (the line labeled 85 μ) and the one-dimensional theory as the two are coincident for the range of burning rates available for testing.

Figure 4.22 presents the data for the bimodal (90 and 600 μ) propellant A-149. Since the large particles of AP tend to decrease the burning rate, it is to be expected that the layer frequency concept (for the 90 μ AP) and the one-dimensional theory are mutually applicable for this propellant. Apparently the layer frequency corresponding to the 600 μ AP was so low that it was unobservable.

The one-dimensional theory parabola deviates quite significantly from the experimental results for the A-151 propellant (45 and 200 μ) as shown in Fig. 4.23. As was mentioned earlier for a burning rate of 0.10 in/sec, the thermal wave thickness for the propellant is approximately 45 μ which coincides with the fine oxidizer particle size for this propellant. Hence, non-one-dimensional behavior was expected for this range of conditions. It should be noted here as well as with the other propellants that the range of conditions for which data may be obtained is very limited in an L^* burner. At low burning rates where the steady-state thermal wave penetrates deeply into the solid there are ignition

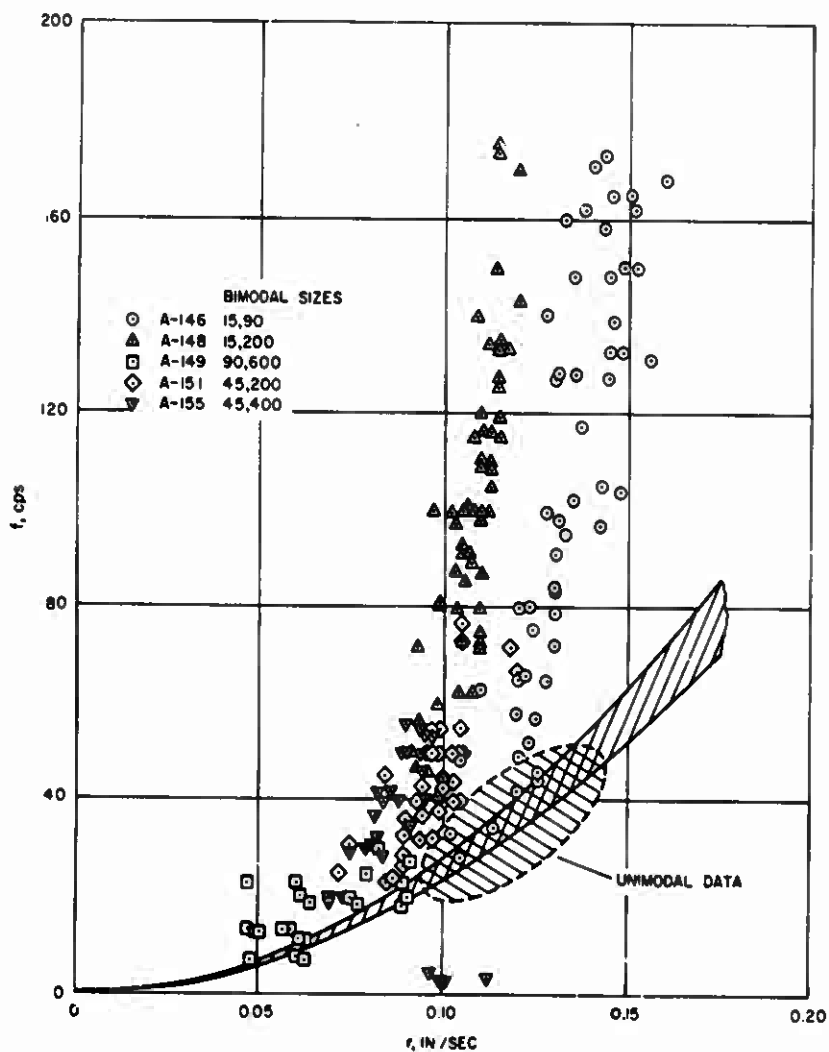


FIG. 4.20. Frequency Versus Burning Rate for the Propellants Tested. The data for the reference, unimodal propellant are indicated by the shaded area and the predicted behavior (based on one-dimensional models) is included.

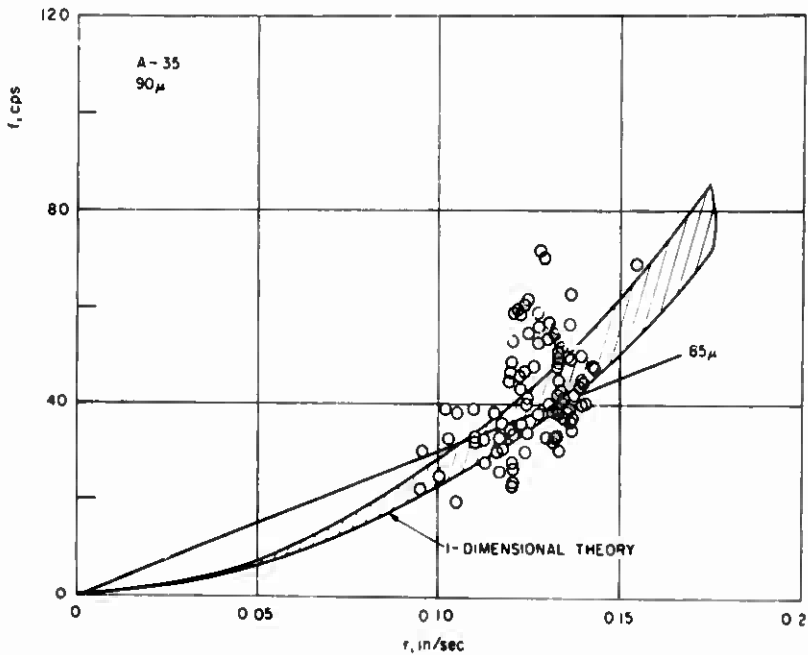


FIG. 4.21. Frequency Versus Burning Rate for the Reference Propellant, A-35. The straight line represents the layer frequency concept based on the median of the particle distribution of the AP.

problems in establishing this profile and igniting the propellant. Also, there are similar problems as the low pressure deflagration limit is reached. At high burning rates, data acquisition is restricted by the stability limit of the L^* burner, and by the physical limit of the burner (i.e., the smaller values of L^* that would be below the stability limit are physically impossible to attain in the burner).

Figures 4.24 and 4.25 are quite similar as might be expected, due to the similarity in the propellant compositions. Both contain 15 μ AP and both oscillated at frequencies two to three times what normally would have been expected. It appears as though there is a transition occurring from one-dimensional behavior at low burning rate to a layer frequency behavior at the higher burning rate.

In Fig. 4.26, propellant A-155 not only exhibits the higher trend in frequencies but also, under certain conditions, oscillated at very low frequencies. The low frequency corresponds very closely to the

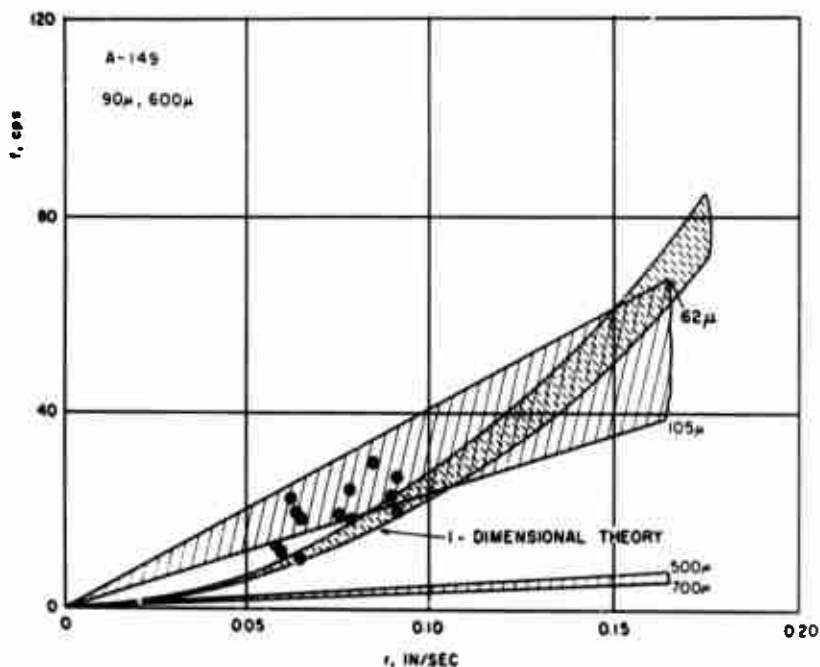


FIG. 4.22. Frequency Versus Burning Rate for A-149.

layer frequency of the larger oxidizer particles. This represents the dual frequency nature that was sought at the outset of the study. Apparently a sufficient number of the large oxidizer particles were burning in phase with each other to cause an oscillation of sufficient magnitude that it could be measured. Figure 4.27 is a pressure-time trace of one of the runs where the low frequency oscillation occurred. The chuffing end oscillations that occur at the initial part of the test are at a frequency of 52 cps while the low amplitude oscillations later in the test are at a frequency of 2.7 cps. This is the most conclusive evidence available that the layer frequency concept is valid and produces an observable effect under certain conditions. Subsequent test records have also exhibited this dual frequency phenomenon.

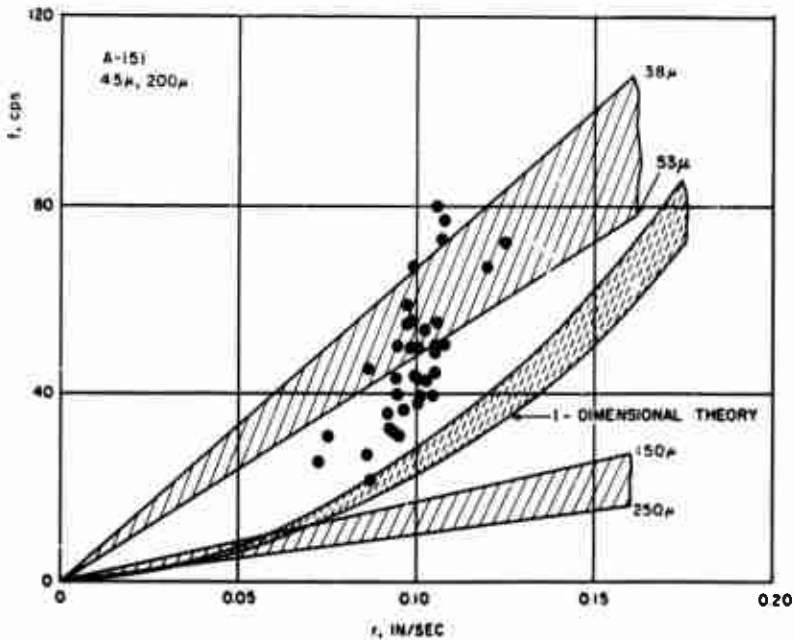


FIG. 4.23. Frequency Versus Burning Rate for A-151.

4.6. THE RELATIONSHIP BETWEEN THEORETICAL MODELS AND THE LAYER FREQUENCY CONCEPT

In order to put the results of this study in perspective with Section 4.3, the data have also been reduced to give the response function and the nondimensional frequency. Figure 4.28 contains the results of the data for four of the bimodal propellants and compares these with results obtained using the Denison and Baum model. The parameters used in the theoretical calculation were determined for A-35 propellant in the manner outlined in Section 4.3 and also by the method recently proposed by Beckstead and Culick (Ref. 30). Utilizing this latter technique to determine the parameters necessary to fit the data for the bimodal propellants results in parameters that are totally impractical (i.e., $A < 0$). This again indicates that these propellants oscillate in a manner that cannot be predicted from one-dimensional theories.

Another way of looking at the data is in a stability plot similar to Fig. 4.15. Utilizing the data for A-155, which seemed to show the most dramatic dual frequency results, Fig. 4.29 was constructed. The data for $\alpha > 0$ should lie to the right of the stability limit line, the

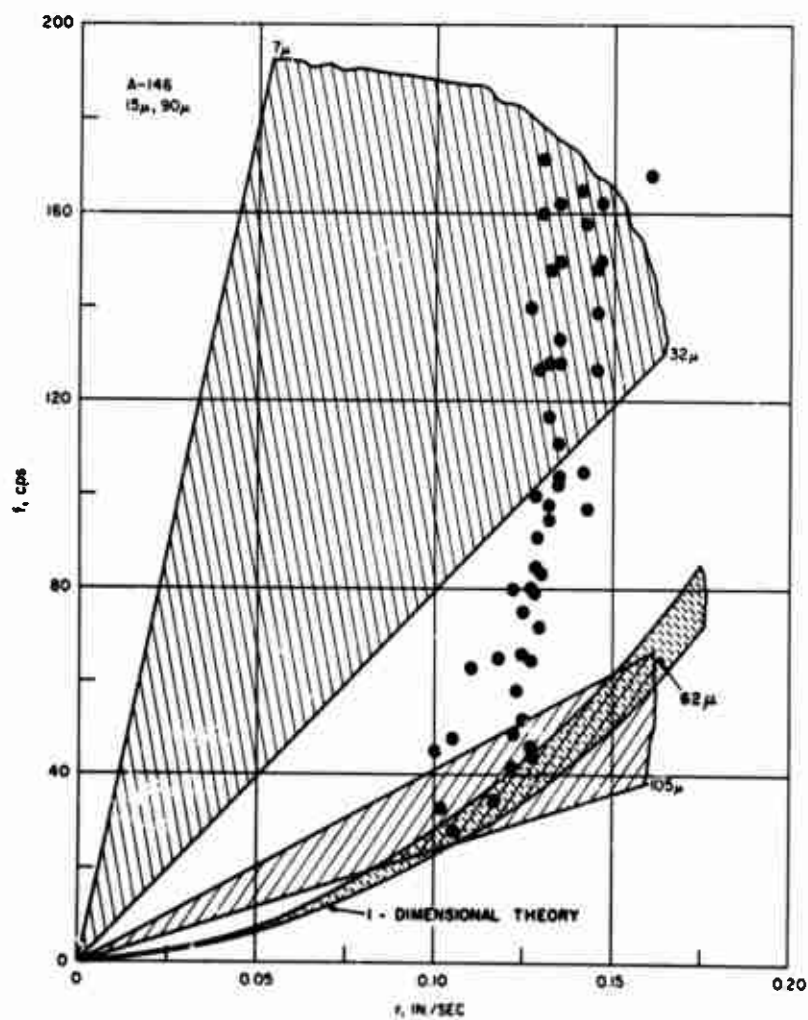


FIG. 4.24. Frequency Versus Burning Rate for A-146.

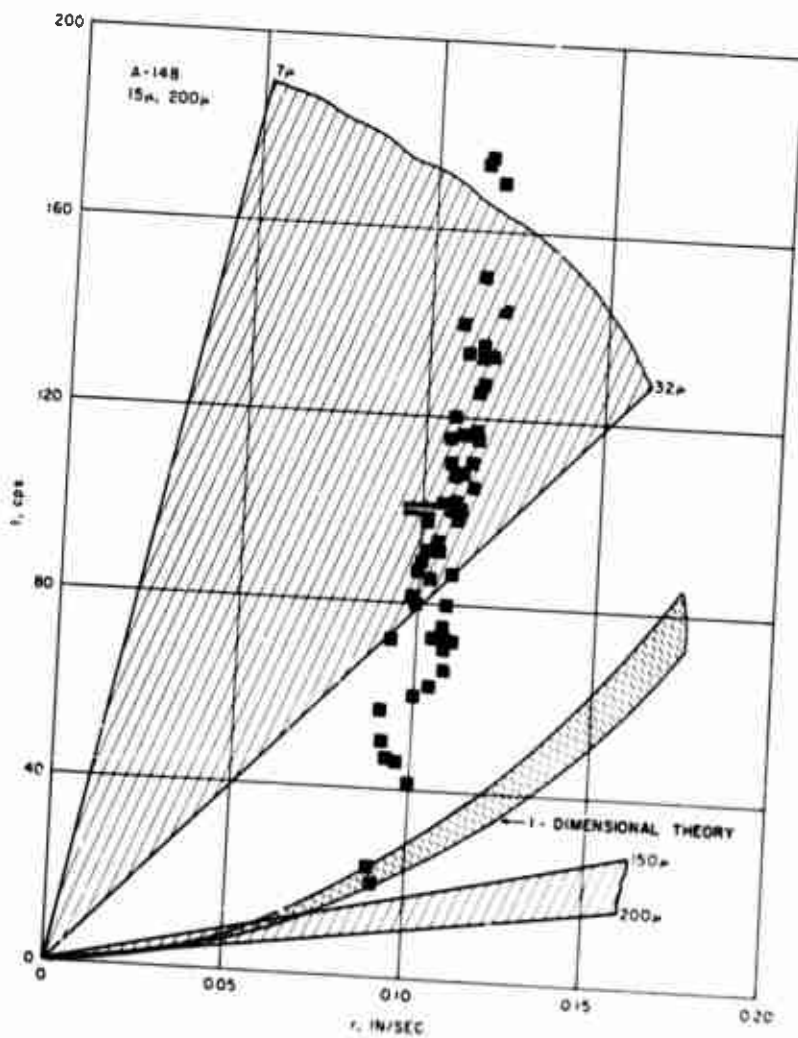


FIG. 4.25. Frequency Versus Burning Rate for A-14B.

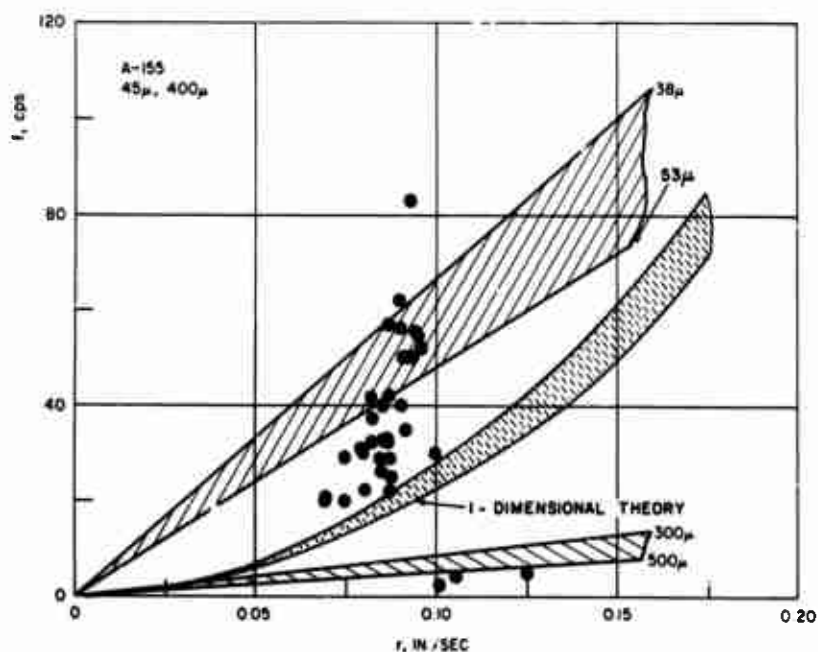


FIG. 4.26. Frequency Versus Burning Rate for A-155.

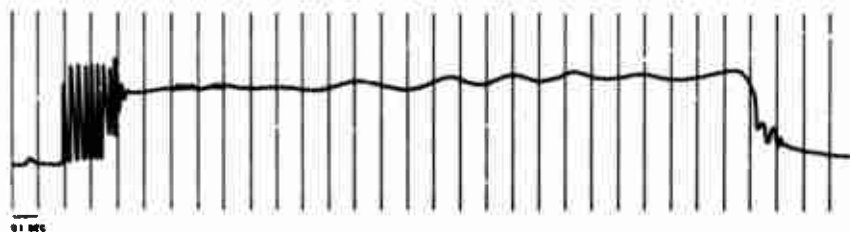


FIG. 4.27. Oscillograph Trace for Propellant A-155 Showing Dual Frequency.

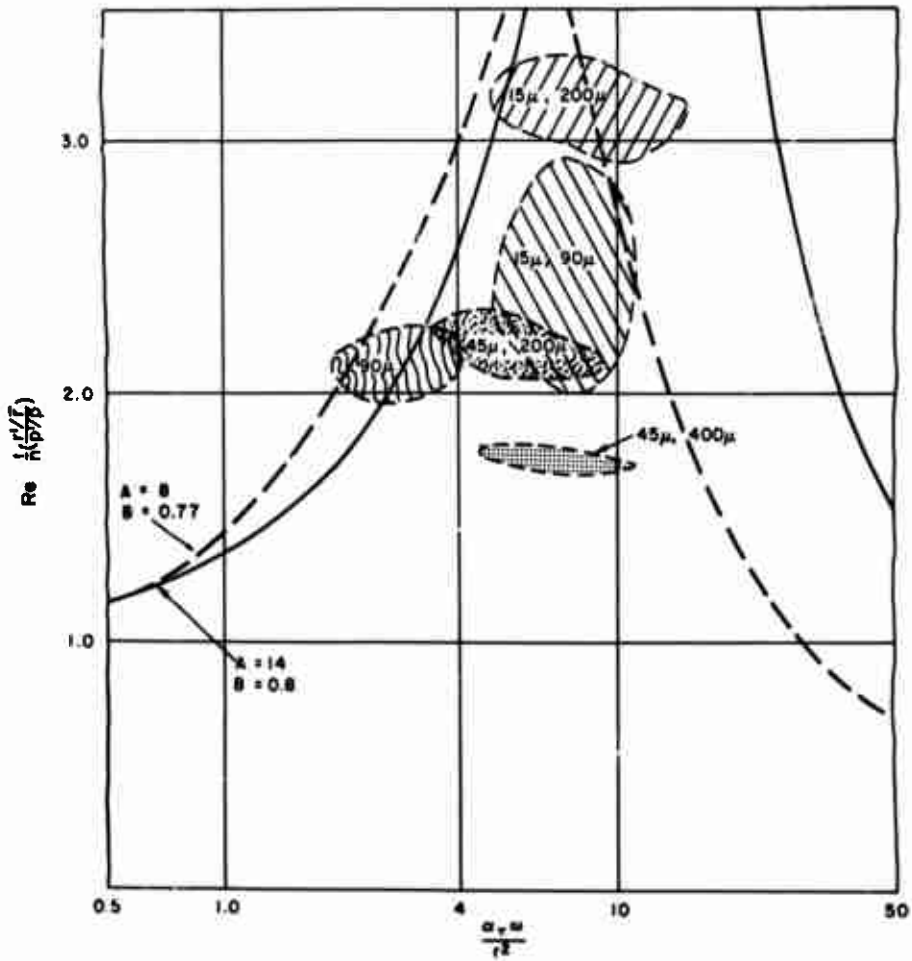


FIG. 4.28. Response Function Versus Nondimensional Frequency.

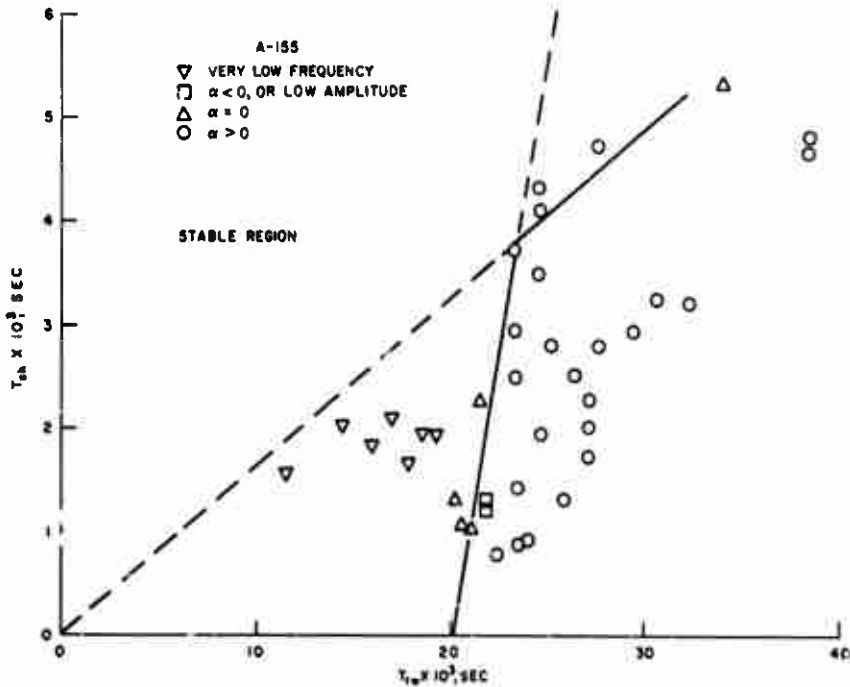


FIG. 4.29. Stability Limit Plot of τ_{ch} Versus τ_{tw} for A-155. Data to the right of the lines are in the unstable region.

$\alpha = 0$ points should lie on the line, and the $\alpha < 0$ points should be to the left of the limit in the stable domain. In the figure all of the points with $\alpha > 0$ lie to the right of the solid line that intersects the abscissa at $\tau_{tw} = 20$ milliseconds. The majority of the data lying to the left of this line are the data at very low frequency. This seems to indicate the possibility of two different stability limits, one corresponding to the very low frequency data and the second for the remainder of the data. It will be recalled from Section 4.4 that the theories predict a straight line through the origin as the stability limit. In Fig. 4.29 the very low frequency data seem to give a stability limit that would agree with theories and yet the frequencies of the oscillations are much lower than would be predicted. This anomalous behavior needs further investigation in order to understand the relation between, and the relative effect of, the lower frequency and one-dimensional theories.

From the above discussion it is apparent that the one-dimensional theories available at present are not adequate to predict the unstable behavior of propellants in a quantitative manner. Nor is it possible to do so with a simple mechanistic argument such as the layer frequency concept. However, by combining aspects of each of these a greater understanding of the problem may possibly be attained. In the following discussion it should be kept in mind that the layer frequency concept results in a range of frequencies under which oscillations occur but gives no indication as to the magnitude of the oscillations, whereas the one-dimensional models give both the frequency and an indication of the magnitude of the oscillations (in the form of the response function). Therefore, the two concepts cannot be combined in an exact sense, but a qualitative combination of the results is very informative. A plot similar to Fig. 4.20-4.26 has been constructed in which the results of the one-dimensional models can be readily included. Figure 4.30 is a log-log plot of the burning rate and the frequency with the layer frequency lines corresponding to A-155 propellant included as well as the line representing the maximum in the response function for $A = 14$ and $B = 0.8$ (see Fig. 4.28). If a third axis extending outward from the plane of the paper is considered to represent the real part of the response function, then Fig. 4.30 takes the form of a contour map. On the map there will be three ridges, two corresponding to the layer frequencies, and the third corresponding to the maximum in the one-dimensional theory. By taking a section of the contour at a constant burning rate ($r = 0.09$ in/sec), a modified form of the response function can be postulated as in Fig. 4.31. The dashed lines represent the approximate effect that could be caused by the layer frequency, and it has been assumed that the two effects would be additive. The magnitude of these ridges due to layer frequency effects are unknown before a series of tests are conducted, but Fig. 4.32 may be used to obtain the frequency that might be expected, given the oxidizer particle size and a particular burn rate. This figure also shows how this result would differ from that expected from a simple one-dimensional analysis, such as the Denison and Baum model.

4.7. SUMMARY

Two prevalent analytical models of solid propellant combustion instability which are relevant to the low-frequency regime have been reduced to a common notation and found to be numerically identical. Non-acoustic data for several propellants have been reduced to a format common with the models, thus allowing a quantitative comparison between theoretical analyses and experimental data of this nature. The possibility of utilizing combustion dynamics models to predict nonacoustic instability behavior has been explored; only qualitative agreement between experimental data and model predictions has been found, and even then several large discrepancies have been discovered. One such discrepancy, the effect of oxidizer particle size, has been explored in detail. A very

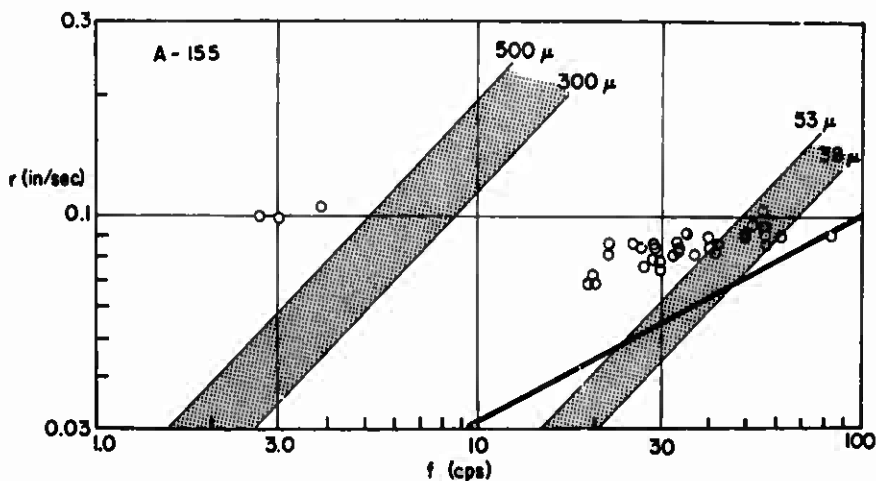


FIG. 4.30. Burning Rate Versus Frequency for A-155 Propellant With Shaded Areas Representing the Layer Frequency Regimes. The heavy solid line is the maximum of the response function calculated from the Denison and Baum model.

simple, phenomenological argument relating frequency of oscillation to the particle size of the oxidizer has been postulated and experimentally examined. This was accomplished by formulating a series of propellants containing bimodal oxidizer blends and testing them in the L^* burner. The results obtained can only be explained by considering aspects contained in the models as well as aspects of the layer frequency concept which is related to the oxidizer particle size.

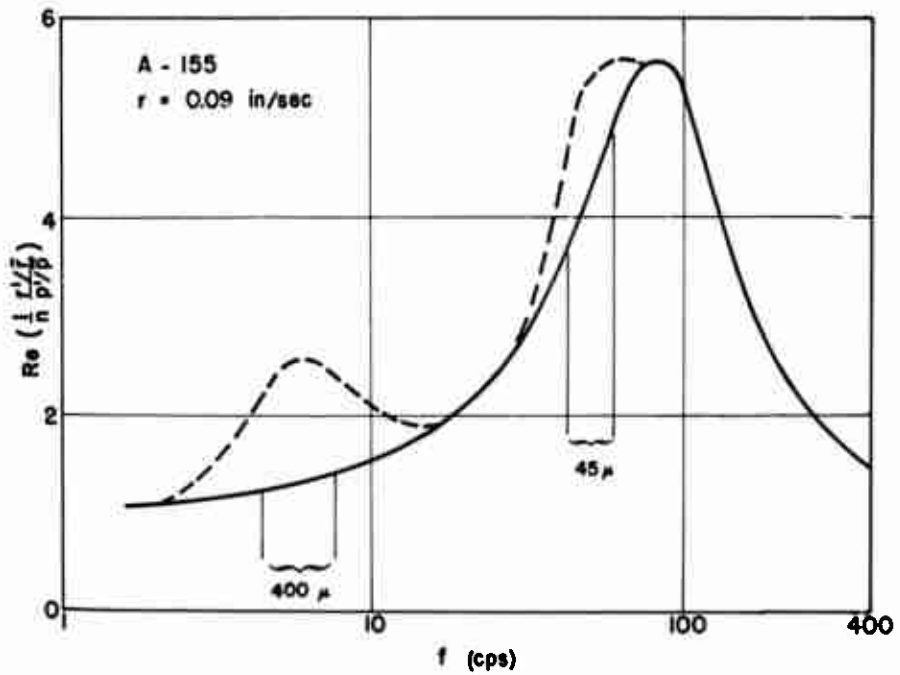


FIG. 4.31. The Modified Response Function With Deviations Included (the Dashed Lines) to Account for Oxidizer Particle Size.

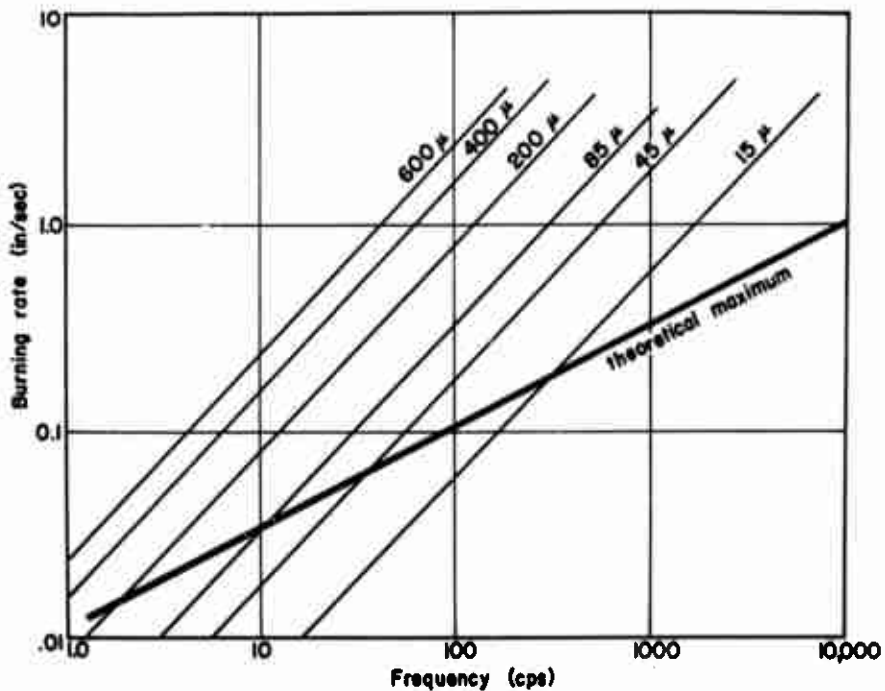


FIG. 4.32. Burning Rate Versus Frequency With the Layer Frequency Lines Included for Typical AP Sizes and the Theoretical Maximum Calculated From a Denison and Bsum Type Model.

5. ACOUSTIC COMBUSTION INSTABILITY

Experimental equipment has been devised which permits the investigation of acoustic instability in the low frequency range characteristic of very large rocket motors (Ref. 31, 32, 33 and 34) and recent modifications to the equipment permit control of the experimental parameters and acquisition of quantitative information including acoustic response function data (Ref. 1). A cutaway diagram of the burner used in this investigation is shown in Fig. 1.

Activity during the present report period included assessment of stability characteristics of several metallized and nonmetallized propellants, initial tests to determine the acoustic response function of a nonmetallized polyurethane propellant composition, tests with pressed AP-Al pellets which indicate that these spontaneously unstable formulations will couple with and drive acoustic wave modes, and an investigation of nonisentropic phenomena in low frequency acoustic instability of solid propellants.

5.1. TESTS TO DETERMINE RANGE OF UNSTABLE BEHAVIOR

Tests were conducted to determine unstable combustion behavior of two compositions for big booster applications (ANB 3105 and ANB 3254), a metallized and a nonmetallized AP-polyurethane propellant (A-147 and A-35), and a nonmetallized AP-PBAN composition (A-13). Results of the big booster propellant tests are discussed in Appendix B.

An investigation previously conducted with a family of metallized AP-polyurethane propellants (Ref. 1) to determine the range of unstable behavior was expanded to include a new composition. Previous tests on A-139 and A-91 propellants revealed that A-139, although unstable over a much wider range of pressure and frequency than A-91, was much harder to ignite (information on propellant compositions appears in Table 5.1). Since the AP particle size was the only parameter varied between A-139 and A-91 it was expected that a propellant with an intermediate oxidizer size might offer a wider range of instability than A-91 and at the same time be easier to ignite than A-139. A 50-pound mix of A-147 was made using 45 μ spherical AP. Tests to determine the range of instability were conducted and the results are shown in Fig. 5.2 with A-139 and A-91 included for comparison. The range of instability for A-147 appeared to be less than that for A-91 and ignitability of A-147 did not appear to be greatly improved over A-139; hence, further tests with A-147 are not planned.

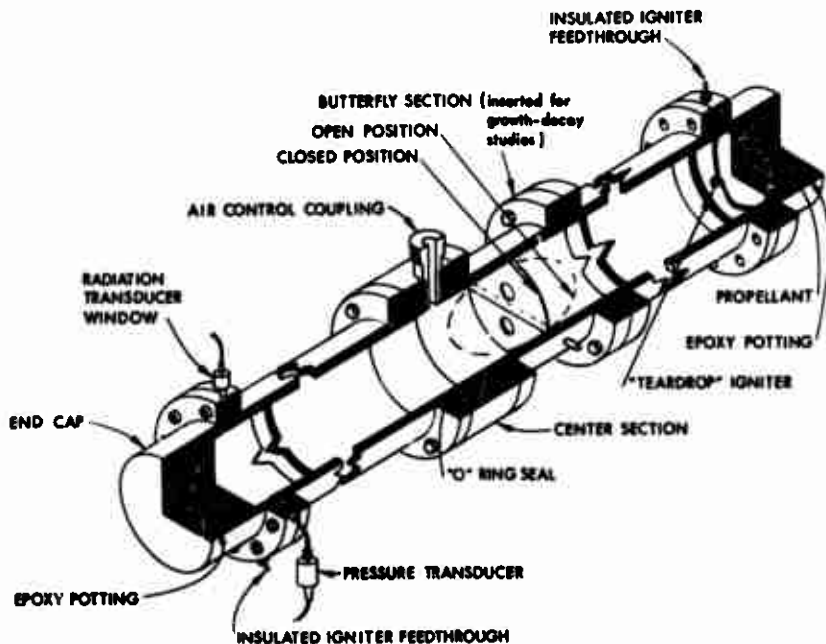


FIG. 5.1. Cutaway Diagram of the 5.5-inch T-Burner.

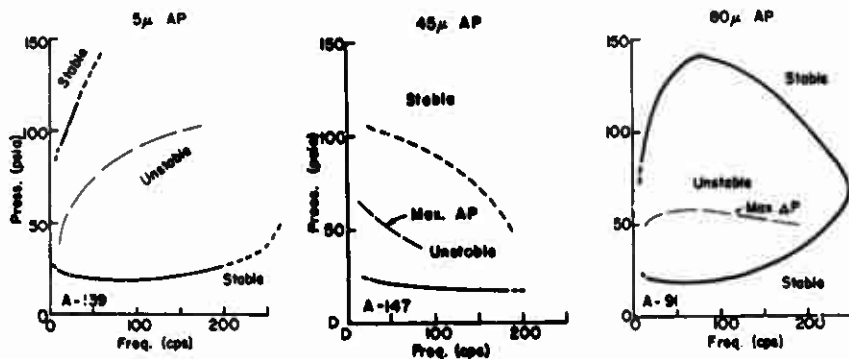


FIG. 5.2. Range of Unstable Combustion Behavior for A-147 Propellant. A-139 and A-91 compositions are included for comparison. Dashed borders indicate uncertain data while conditions of maximum acoustic pressure are indicated by the broken line in the unstable area.

TABLE 5.1. Compositions Used in Low Frequency Acoustic Instability Research

Designation of composition	Binder		Oxidizer (spherical AP)		Aluminum (spherical)		Other	
	Type	Wt %	Mean dis.	Wt %	Mean dis.	Wt %	Type	Wt %
A-13	PBAN	24.0	80 μ	76.00	--	--	--	--
A-35	Polyurethane	25.0	80 μ	75.00	--	--	--	--
A-91	do	25.0	80 μ	67.00	5 μ	8	--	--
A-139	do	25.0	15 μ	67.00	5 μ	8	--	--
A-147	do	25.0	45 μ	67.00	5 μ	8	--	--
A-152	do	25.0	80 μ	74.60	--	--	NaCl	0.4
PU	do	20.1	5 μ	23.85	--	--	NaCl	0.05
			45 μ	56.00	--	--	--	--
J-30	--	--	80 μ	94.00	5 μ	4	CuO2O2	2.00
J-81	--	--	80 μ	88.00	15 μ	4	CuO	8.00

Two unmetallized propellants, A-13 and A-35 (Table 5.1), which have been extensively tested in the 1.5-inch T-burner, were chosen for study in the low frequency range in the 5.5-inch burner. Testing of these two compositions to date has been primarily to determine the range of unstable combustion behavior. Results are shown in Fig. 5.3.

A notable feature of tests with A-13 was the simultaneous presence of several axial acoustic modes during tests when the fundamental mode frequency was below 90 cps. Validity of the response function determination depends on presence of a single acoustic mode so that A-13 response function tests appear to be limited to frequencies above about 90 cps. Tests with A-35 over a range of frequencies similar to the A-13 tests did not reveal presence of modes higher than the fundamental axial mode and there does not appear to be a combustion-imposed lower frequency limit as with A-13.

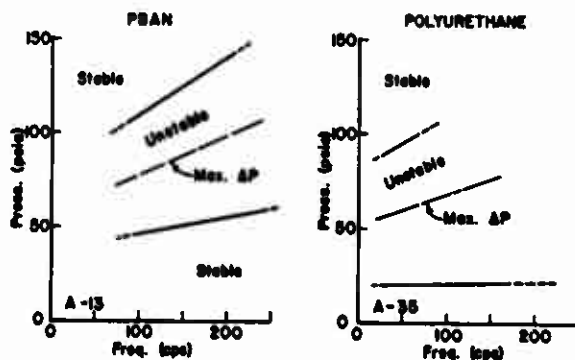


FIG. 5.3. Range of Instability Behavior for Two Unmetallized Propellants.

5.2. PRESSED PELLET INSTABILITY

Spontaneous combustion oscillations observed with AP-Al pressed pellets have been previously reported (Ref. 1). Investigations with metallized propellants which appeared to have a preferred frequency behavior have also been published (Ref. 35), but the correlation of a preferred frequency characteristic with metal combustion in propellant composition has been indirect. Thus, pressed AP-Al materials appear to offer a means for conducting a definitive study of the relationship between metal combustion and preferred frequency combustion oscillations.

The relevance of pressed pellet combustion oscillations to acoustic instability has not been tested until recently. At least two aspects are involved which determine whether spontaneous combustion oscillations will excite an acoustic wave mode oscillation: (1) whether the combustion oscillations generate a pressure perturbation, and (2) whether the combustion oscillations are capable of being coupled with flow disturbances in a manner which will result in amplification of an acoustic mode (Ref. 36).

Initial tests to determine the extent to which pressed AP-Al materials exhibit acoustic instability have been conducted with J-30 (see Table 5.1) in the 5.5-inch burner. The problems of attaining large pellet size and of mounting the pellets without the use of organic binders or potting compounds which inhibit spontaneous instability, were solved by utilizing an existing die of approximately 1-1/2-in. diameter and arranging the pellets in an array such that seven pellets were accommodated in each end of the burner. Mounting was accomplished by pressing the pellets into a metal disc (Fig. 5.4). The metal disc-pellet assembly was then held in place in the bottom of the burner end-cap by a backing of silicone-glass adhesive tape. Ignition was accomplished by use of the same paint-on material used for propellants.

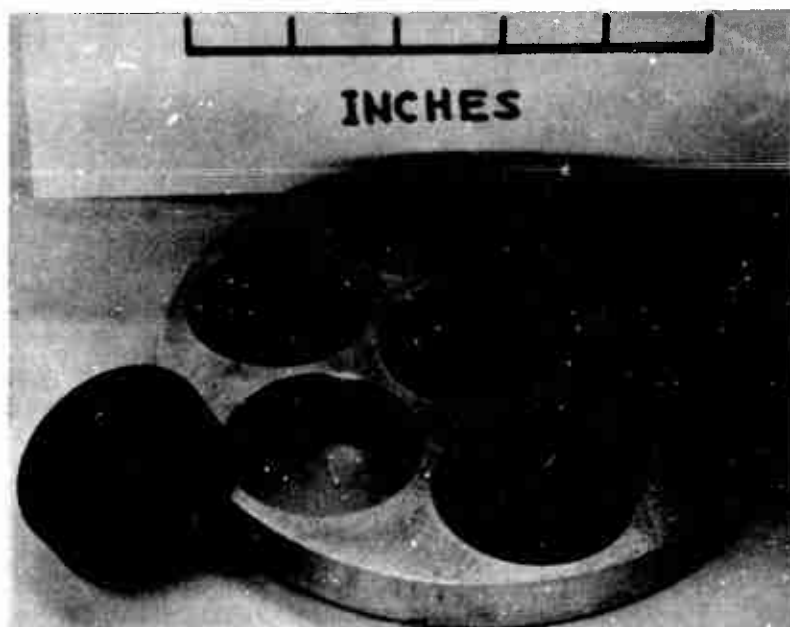


FIG. 5.4. Pressed Pellet Mounting Arrangement for the 5.5-inch Burner.

Test results indicated that coupling did occur and that the pressed pellet material is a good medium for study of the role of metal behavior in producing preferred frequency instability. Further testing, however, will be more readily accomplished by choice of a pressed-pellet formulation that exhibits spontaneous oscillatory behavior at lower pressures than J-30. J-81 (Table 5.1) appears to be a suitable candidate, as noted in Ref. 1.

5.3. INVESTIGATION OF NONISENTROPIC PHENOMENA

5.3.1. Introduction

Knowledge of the response of propellant combustion processes to flow perturbations is vital to any quantitative description of acoustic instability. The acoustic admittance appears as a boundary condition in solution of the wave equation and it describes the flow of energy into or out of the acoustic wave. The admittance is defined as

$$Y \equiv - \frac{v'}{p'} \quad (5.1)$$

where v' is a velocity perturbation normal to the propellant surface and p' is a pressure perturbation. When written in terms of variables related to propellant combustion, the admittance becomes (Ref. 34)

$$Y = - \frac{\bar{v}}{\bar{p}} \left\{ \frac{\mu}{\epsilon} - \frac{\sigma}{\epsilon} \right\} \quad (5.2)$$

where \bar{v} is the mean gas velocity, \bar{p} is the mean pressure, μ/ϵ is the ratio of mass perturbation to pressure perturbation, and σ/ϵ is the ratio of density to pressure perturbation. It is assumed that the above quantities are calculated at a plane near the propellant surface but at a location outside the chemical reaction zone. It is further assumed that gas velocity and pressure fluctuations are described by a standing acoustic wave mode, that the flame zone is of a thickness small compared to other dimensions in the system, and that the gas-phase reaction times are short compared to a period of acoustic oscillation and to the transit time of an element of gas through the combustion zone. It should be noted that, in general, Y is a complex quantity. However, the real part of the complex admittance determines whether or not an acoustic wave will be amplified; the out-of-phase (or imaginary) component of the combustion response cannot amplify a pressure perturbation.

The ratio μ/ϵ is commonly referred to as the "response function" and considerable theoretical and experimental investigation has been devoted to determining its magnitude. Values of the response function, according to T-burner results, typically fall in the range between unity and 5. Less emphasis has been put on evaluating σ/ϵ since theoretical studies indicate that it is of the order of unity in magnitude. It is this ratio which is sensitive to the nature of the gas processes in the combustion zone. If the combustion zone behaves isentropically, $\sigma/\epsilon \rightarrow 1/\gamma$, while it approaches unity if the combustion zone behaves isothermally. This latter situation is often referred to as the "zero-frequency case". Actual values of this ratio probably lie between these two extremes, varying according to conditions.

In spite of the reduced importance of σ/ϵ to determination of the admittance, interest in experimental investigation of conditions under which nonisentropic behavior appears has persisted. Attempts to verify nonisentropic behavior during acoustic instability have, until the present, been negative or inconclusive. In theory the isothermal case assumes that the flame temperature in an oscillating environment is constant. Under typical rocket motor operating conditions the flame temperature is relatively insensitive to pressure so the above assumption would appear to be reasonably valid. Under these conditions, successive elements of gas enter the acoustic field with different values of entropy which is given by the equation (Ref. 37)

$$s = s_0 + \epsilon R(\sin \{\omega t - \tau\}), \quad (5.3)$$

where a_0 is a reference value of entropy, ϵ is the ratio of maximum pressure fluctuation to mean pressure, R is the gas constant, ω is the angular frequency of the acoustic wave, and t represents time.

Entropy variations are detected as temperature fluctuations given by the relation

$$T(x,t) = T_f \left\{ 1 + 2\epsilon \left(\frac{\gamma-1}{\gamma} \right) \sin \frac{\omega x}{2v_g} \cos \left(\omega t - \frac{\omega x}{2v_g} \right) \right\} \quad (5.4)$$

where T_f is the flame temperature, x is the distance from the propellant surface, and v_g is the gas velocity (Ref. 37)

It was noted in Ref. 37 that at any instant in time there would not only be a sinusoidal temperature variation in space, with amplitude $T_f \epsilon (\gamma-1)/\gamma$, but that there would also be a time-varying temperature observed at fixed points on the burner. In addition, if one were to observe the magnitude of temperature fluctuations at various points along the burner, it would be found that the amplitude of temperature fluctuations varies with distance from the propellant surface. Maxima in the temperature fluctuation would be found at locations given by

$$x_{T, \max} = (n + \frac{1}{2}) v_g T, \quad (5.5)$$

while the temperature fluctuation would be zero at points located at

$$x_{T, \min} = n v_g T, \quad (5.6)$$

where n represents any positive integer and T is the period of acoustic oscillation. The amplitude of temperature fluctuation (Eq. 5.5) is equal to $2 T_f \epsilon (\gamma-1)/\gamma$, i.e., twice the value of the spatial temperature variation.

Equations 5.4, 5.5, and 5.6 provide a standard for comparing the foregoing theory with experimental observations. The observation of gas temperature variations as a function of space and/or time coupled with knowledge of the acoustic frequency and the gas velocity should be sufficient to verify existence of the entropy wave phenomenon. Sufficiently detailed knowledge of the relationship between the temperature fluctuations and the acoustic pressure would also permit a determination of the o/c term through the "temperature response function" as discussed in Ref. 37 and 38.

Several attempts have been made in the past to detect nonisentropic behavior but these efforts did not yield a positive result. Recent

theoretical studies indicated that the previous experiments may have failed because they were conducted at frequencies ill-suited for displaying of the phenomenon (Ref. 39). Since the NWC 5.5-in. burner was capable of operating within the frequency range of interest, it was decided to attempt to obtain evidence of nonisentropic behavior in that apparatus.

Two types of experiments were conducted: (1) a series in which photographic recording was utilized to obtain space-time records of the light intensity variations in the gas after it left the combustion zone; (2) a series in which high-speed temperature measurements were made using an adaptation of the brightness-emissivity technique. Three propellant compositions were used in the two series of experiments: (1) a bimodal polyurethane-AP composition prepared at Princeton University (designated PU in this report), and cast into metal cups, (2) a modified version of A-35 containing 0.4% NaCl which was designated A-152, and (3) A-13 propellant. Composition of these propellants appears in Table 5.1.

5.3.2. Photographic Studies

The series of photographic experiments utilized a special burner test section which was fitted with a nitrogen-flushed slit window (Fig. 5.5). An optical bench was used to mount a 70 mm streak camera and a long focal length objective lens. A cathode ray oscilloscope and mirror were arranged so that the oscilloscope screen was focused on the film plane in the camera in line with the image of the window slit image. Thus the acoustic pressure trace, which appeared on the cathode ray screen, was recorded simultaneously with the radiant intensity variations that appeared in the slit window on the burner. This arrangement permitted a detailed comparison to be made between events occurring in the burner gas and the acoustic pressure. A block diagram of the equipment arrangement used in the photographic studies is shown in Fig. 5.6.

A portion of streak camera record is shown in Fig. 5.7. The acoustic pressure and approximate position of the propellant surface have been added to the figure as an aid in interpretation. Major features of interest have been inked onto the figure to the left of the photographic record for the same reason. The photographic portion of this figure is a positive so that high and low temperature regions in the figure appear as light and dark areas respectively. Calculations from Eq. 5.5 and 5.6, based on an estimate of the mean value of the gas velocity as it leaves the propellant surface, were made in order to locate the positions of the maximum and minimum temperature variations. These are designated in the figure as T'_{\max} and T'_{\min} respectively. Note also that a time and distance scale are provided in Fig. 5.7 and that time runs from right to left.

Major features in the reconstructed portion of the record are identified with letters as follows:

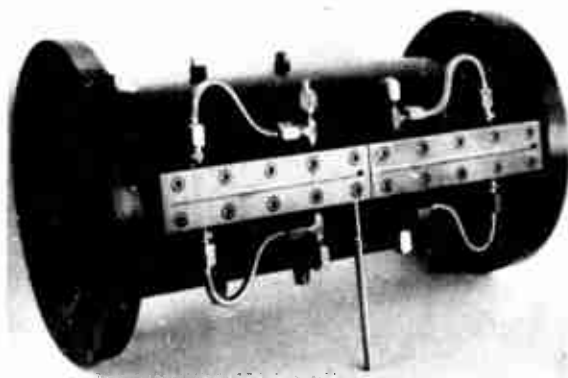


FIG. 5.5. Test Section Containing Nitrogen Flushed Slit Window Used on the 5.5-Inch T-Burner for Photographic Studies of Nonisentropic Phenomena in Low Frequency Acoustic Instability.

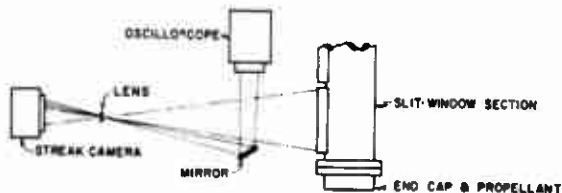


FIG. 5.6. Block Diagram of Streak Camera Arrangement Used for Photographic Study of Nonisentropic Phenomena in the 5.5-Inch Diameter Burner.

A. Periodic vertical darkened zones appear in the record which occur at times when the acoustic pressure is a minimum. These areas appear to be related to adiabatic cooling of gas throughout the burner section covered by the window caused by lowering of the acoustic pressure. (Note that there is also a general brightening at acoustic pressure maxima related to adiabatic heating from compression of the gas in the burner by the acoustic wave.)

B. Dark diagonal lines, designated "B", are relatively persistent in their travel across the section covered by the window. These represent elements of relatively cold gas which appear to have originated from the propellant surface at a time when the acoustic pressure was a minimum. This assertion of the origin of these elements of gas is

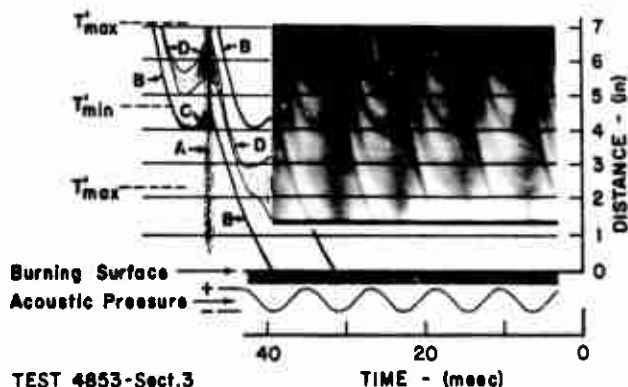


FIG. 5.7. Portion of Streak Camera Record from Test with PU Propellant. Mean burner pressure was 75 psia, acoustic pressure amplitude was 9 psi p-p, time after ignition was 2.3 sec. The position of the burning surface, the acoustic pressure, and some of the major features of the record have been reconstructed for clarity. Points of particular interest in the reconstructed portion of the record are designated by letters in the figure, with appropriate description in the test. Locations of maximum and minimum temperature fluctuations, designated T'_{max} and T'_{min} respectively, were calculated from Eq. 5.5 and 5.6.

based on an extrapolation of the information on the photographic record back to the propellant surface as indicated in the inked reconstruction to the left of the photographic portion of the figure. It was assumed in making the extrapolation that the gas velocity in the region of extrapolation is relatively unaffected by the acoustic wave. If the interpretation given here is correct, a phenomenon is present in these tests that the theory in Section 5.3.1 does not take into account.

C. Apparent inflection points occur in the motion of cold gas elements (designated "C"). The photographic record suggests that a significant fluctuation occurs involving, perhaps, a momentary flow reversal. It should be noted that the theory described in Section 5.3.1 assumes uniform gas flow and does not provide for a flow perturbation of the sort indicated in the record.

D. Dark areas designated "D" represent regions of cold gas, which at any instant in time occupy a position midway between "B" elements of gas. Since these appear between areas of gas ("B" designation) which originated at acoustic pressure minima, it seems that the "D" regions are related to the entropy fluctuations referred to in Section 5.3.1.

There are several difficulties related to the above discussion. First is the problem of identification of the cold gas that appears to leave the propellant surface at an acoustic pressure minimum. The extrapolation of an element of gas back to the surface has to be done over more than an inch for which there is no photographic record. In doing the extrapolation, the assumption has been made that the acoustic velocity perturbation in this region is negligible since it is so close to the propellant surface, relative to the acoustic wavelength. Second, the film was overexposed causing a diminishing of the relative importance of cooler regions in the gss. Some of the effects of overexposure have been counteracted by printing on high-contrast material but this may have introduced further distortion of the quality of the photograph. Problems in photographic record interpretation can probably be resolved by repeating tests with altered burner parts which allow the propellant to be moved up to the end of the window and by using a camera lens with smaller aperture than was used in the first test series.

5.3.3. High-Speed Temperature Measurements

The second part of this investigation involved acquisition of high-speed gas temperature data from a region near the propellant surface. The measurement technique utilized an adaptation of the brightness-emissivity approach mentioned in Ref. 37. The apparatus is shown in Fig. 5.8 and consists of: (A) a ribbon filament lamp of known brightness temperature, (B) a system of lenses and slits to focus and collimate the light, (C) a rotating perforated metal wheel which periodically interrupts light from the known source, (D) a special test section which fits between the burner and an end cap containing propellant that contains two small nitrogen-flushed windows which permit light from the known source to pass through the burner, and (E) a monochromator equipped with a photomultiplier tube which receives and detects radiation in the sodium D-line portion of the optical spectrum. The photomultiplier signal was recorded on an oscillograph simultaneously with the chamber pressure. Prior to each run, the source was calibrated with a precision pyrometer and a short calibration record was obtained on the oscillograph giving the deflection due to the lamp intensity. During the test, the radiation received by the photomultiplier tube alternated between that from the flame and that of the flame plus radiation from the source. A portion of a test record is shown in Fig. 5.9.

Data reduction following a test was accomplished by making deflection measurements of the pre-run source calibration, and measuring the signal deflections from the photomultiplier and the pressure transducer over one or more cycles of oscillation at selected portions of the test. A typical plot of temperature-pressure data is shown in Fig. 5.10.

Data from selected tests appear in Table 5.2. Columns [a] through [h] represent data derived from the oscillograph recordings. Alphabetical suffixes to the test number are used to identify the portion of the test record from which the data was taken. The time at which the data

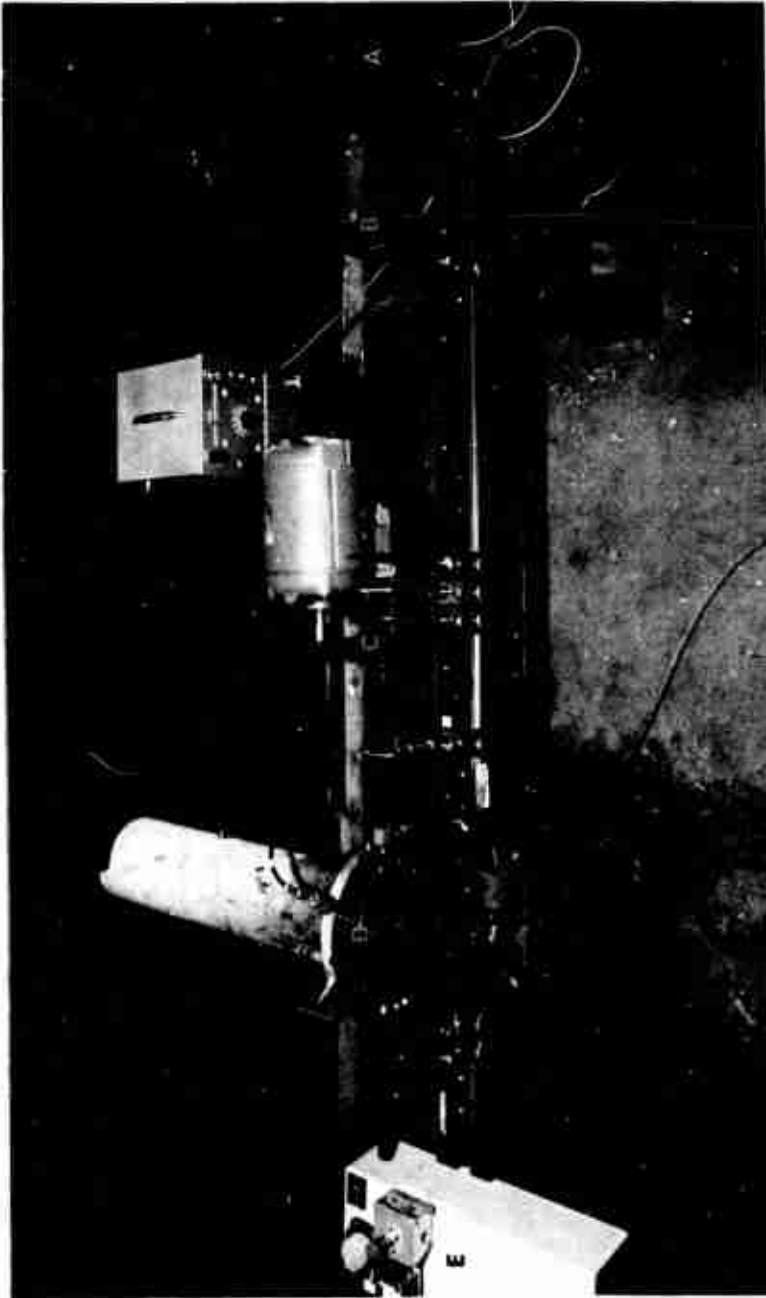


FIG. 5.8. Apparatus Used for Brightness-Emissivity Temperature Determination. Lettered items are described in the text.

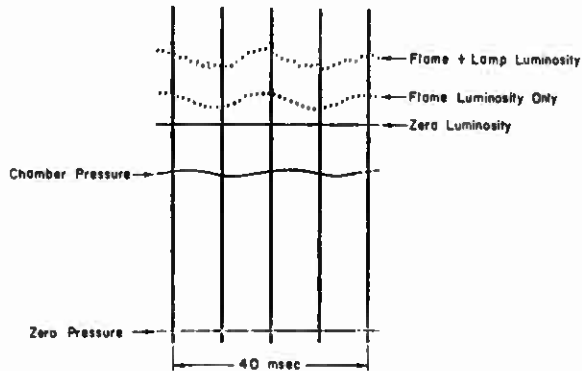


FIG. 5.9. Portion of Test Record From a Test in Which High-Speed Temperature Data was Acquired.

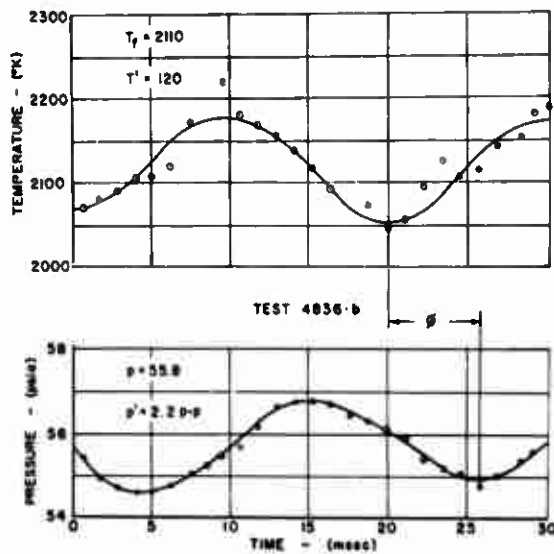


FIG. 5.10. Temperature-Pressure Data Reduced and Plotted on a Common Time Base From Test Record Similar to That Shown in Fig. 5.9.

TABLE 5-2. Pressure-Temperature Data.

Propellant type	Test no.	Time ^a t sec [a]	Freq f cps [b]	Pressure		Temperature		Severity of pressure oscillation ϵ [g]	Phase ^{**} ϕ fraction of cycle [h]	$\frac{T}{\gamma} - 1$ $\frac{\epsilon}{\gamma}$ [i]	S ^{***} °K p-p [j]
				Mean p psia [c]	Oscillation amplitude psi p-p [d]	Mean T °K [e]	Oscillation amplitude °K p-p [f]				
PU	4826A	3.81	15.0	46.5	4.95	2064	44	0.106	0.078	43.8	70.7
	4829B	2.70	37.0	53.7	5.25	2220	100	.098	-.007	43.4	75.0
A-152	4825A	2.30	14.6	44.0	10.0	2008	190	.227	-.415	91.2	48.5
	4825B	2.60	14.6	46.5	9.0	1980	155	.194	-.465	76.8	43.7
	4836C	2.73	46.0	55.95	1.97	2120	120	.039	.026	16.4	9.55
	4836B	3.38	46.0	50.80	2.20	2110	120	.039	.262	16.6	10.67
A-13	4837B	2.26	52.5	68.0	1.23	2130	135	.018	.105	7.70	5.97
	4832A	2.88	81.0	26.7	1.3	2188	100	.049	-.062	21.3
	4832B	3.18	78.7	27.5	2.4	2165	165	.087	-.061	37.8

^a Time at which data was taken.^{**} Phase of temperature fluctuation relative to the pressure fluctuation. Negative sign indicates a temperature lag.^{***} S = sensitivity of temperature to pressure. S = 14.3°K/psi for PU propellant and S = 4.85°K/psi for A-152 propellant.

was taken appears in column [a] and is the time elapsed after ignition. Frequency was determined by measurement of the oscillograph record (column [b]) while the pressure and temperature data were obtained from plots similar to that shown in Fig. 5.10. Column [g] was calculated by taking the ratio of the pressure oscillation amplitude to the mean pressure. The phase between temperature and pressure (column [h]) was computed from measurements of the pressure-temperature plots (as in Fig. 5.10). Columns [i] and [j] are computed temperature fluctuations that will be discussed in some detail below.

In general, the tabulated pressure-temperature data show quite a diversity of behavior. The A-152 data represent the most comprehensive compilation of information available at present, but the question of whether the data fits a consistent pattern must await completion of the data analysis. One outstanding feature, for which there is no explanation at present, is the surprisingly high amplitude of temperature oscillation observed in the tests above 40 cps. A comparison of observed temperature fluctuation amplitude with calculated amplitudes, based on two rather different assumptions suggests that neither of the explanations for the temperature oscillations advanced so far can explain what is observed in the burner. The first comparison is with the temperature fluctuation predicted on the assumption that the gas in the burner behaves isentropically. This assumption leads to the calculated temperature entered in column [i]. Comparison of the isentropic calculation and the observed temperature oscillations indicates the increased difference between the isentropic prediction and the observed values. If the assumptions of Section 5.3.1 were valid, the observed temperature oscillation amplitude would not exceed the isentropic value by more than a factor of two. As one can see, the observed amplitudes from tests with frequencies above 40 cps far exceed the expected range.

Another facet of the temperature oscillations was considered, namely the effect of a fluctuating flame temperature. It was noted, in comparing mean temperature and mean pressure from PU and A-152 data, that the temperature was a function of pressure. The dependence of temperature on pressure for these two propellants is indicated in Fig. 5.11. Assuming that the temperature follows the pressure oscillations in the same manner as indicated in Fig. 5.11, one can assign a sensitivity of temperature to pressure which can be used to compute the temperature oscillation caused by an observed pressure fluctuation. Computations based on the above assumptions are entered in column [j] of Table 5.2 and the temperature sensitivity for the two propellants appears in a footnote in that table. Comparison of the observed and computed data indicates a large disparity and that the temperature sensitivity to pressure does not offer an adequate explanation of the large temperature oscillations observed at the higher frequencies.

A detailed report is in preparation which describes the photographic and high-speed temperature measurements in greater detail. The information will be released as a NWC report when completed.

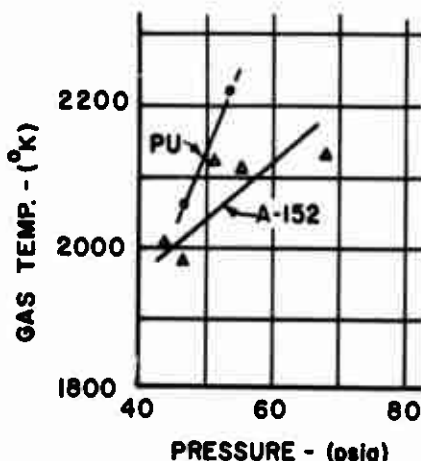


FIG. 5.11. Plot of Measured Mean Gas Temperature and Mean Pressure for PU and A-152 Propellants.

5.3.4. A New Mathematical Model

Detailed interpretation of the streak photography and the temperature data is hampered by presence of phenomena not accounted for in existing theories. It was felt that a mathematical model could be devised which would account for a variable flame temperature and non-uniform flow of gases. Steps in that direction were taken late in the report period.

A new theory is presented here which describes the history of a gas particle from the time it leaves the reaction zone as a function of a parameter ψ which denotes the phasing of the pressure and velocity fields at the emergence of the particle from the combustion zone.

The oscillating components of pressure and velocity were treated as first order perturbation quantities and since the amplitude of the oscillating component of velocity was of the order of the mean velocity, a point clearly shown by the evidence of flow reversal in some of the photographic test records, the nonconvective wave equation was used to describe the acoustic fields. Pressure antinodes at each end of the chamber were used as boundary conditions with the resulting oscillating pressure and velocity fields given by

$$p' = \bar{p} \epsilon \cos (\pi x/L) \cos (\omega t + \psi) \quad (5.5)$$

and

$$v' = \bar{V} \sin (\pi x/L) \sin (\omega t + \psi) \quad (5.6)$$

respectively. In Eq. 5.5 and 5.6 \bar{p} is the mean pressure, ϵ is the ratio of the maximum value of the pressure perturbation to mean pressure, \bar{V} is the velocity amplitude, x is the distance from the reaction zone and L is the T-burner length. The particle velocity is given by

$$v = \bar{V} + v' \quad (5.7)$$

and the relationship between temperature and pressure given by

$$\frac{T'}{T_r} = \left(\frac{\gamma-1}{\gamma} \right) \frac{p - p_r}{p_r} \quad (5.8)$$

in which p_r was the pressure at a reference plane immediately outside of the flame when the particle in question entered the acoustic field and T_r is the temperature at the reference plane, which is assumed to be constant for this discussion. The reference pressure given by

$$p_r = \bar{p} + \bar{p}\epsilon \cos \psi \quad (5.9)$$

was obtained by adding the mean pressure \bar{p} to the acoustic pressure with x and t set equal to zero. The following expression for relative temperature

$$\begin{aligned} \frac{T}{T_r} &= \frac{T' + T_r}{T_r} = \\ 1 + \left(\frac{\gamma-1}{\gamma} \right) \epsilon &\left[\frac{\cos (\pi x/L) \cos (\omega t + \psi) - \cos \psi}{1 + \epsilon \cos \psi} \right] \end{aligned} \quad (5.10)$$

was obtained by substitution of Eq. 5.7 and 5.9 into Eq. 5.8. In order to evaluate T/T_r it is necessary to determine the relationship between x and t for a given ψ . This is readily obtained from the relation

$$x = \int_0^t v \, dt = \int_0^t [\bar{V} + \bar{V} \sin (\pi x/L) \sin (\omega t + \psi)] \, dt \quad (5.11)$$

Solution of Eq. 5.11 thus gives the trajectory of a gas particle leaving the surface at time zero for the assumed boundary conditions and phase angle ψ . Equations 5.10 and 5.11 together give the temperature history and the trajectory of a gas particle subsequent to leaving the surface as a function of the parameter ψ .

Additional steps in modifying the analysis to provide a more adequate description of significant parameters in the problem of nonisentropic behavior need to be undertaken. These include replacing the rigid solid boundary condition at the combustion zone with an acoustic admittance and replacing the constant flame temperature with one which is pressure sensitive.

It is planned to present solutions of the equations in the form of dimensionless plots. With these available, particle trajectories and constant temperature lines may then be superimposed on experimental test records, the phase angle ψ being readily obtained from the pressure record. The relative brightness patterns in the streak data will then provide a qualitative test of the mathematical model.

5.4. SUMMARY

Tests to determine range of unstable combustion behavior on one metallized and two unmetallized propellants were accomplished prior to more definitive testing. It is anticipated that extensive response function testing on the two unmetallized compositions will be accomplished in the near future.

Preliminary testing with a preseed AP-AI composition in the 5.5-inch diameter T-burner demonstrated the feasibility of using such materials in investigating the role of metal combustion in preferred-frequency instability.

Investigation of nonisentropic behavior in the low frequency range revealed that, under the conditions tested, the flame temperature may fluctuate significantly with the acoustic pressure. While it appeared that the flame temperature of two of the propellants tested was a function of the mean pressure, the resulting temperature variation was not large enough in many cases to explain the observed temperature fluctuations. The magnitude of the temperature oscillations and the evidence from the photographic records strongly suggested that factors other than those discussed above were involved. One plausible, but as yet unevaluated, explanation was that the binder and oxidizer were consumed at different rates which vary over a cycle of pressure oscillation, resulting in a fluctuation in the fuel-to-oxidizer ratio.

The observations of nonisentropic behavior strongly suggested that present mathematical models which seek to describe unstable combustion may need additional refinements in order to accurately describe the phenomena that appear to be involved.

6. SUMMARY

Each of the preceding sections contains its own extensive discussion, and the major accomplishments of each section are summarized here in order to provide a comprehensive view of the entire program.

6.1. DECOMPOSITION AND DEFLAGRATION OF AMMONIUM PERCHLORATE

Past and current work has shown that the physical character of the surface residue of AP is significantly dependent on the imposed heating rate. Ammonium perchlorate apparently sublimes under conditions of isothermal decomposition and there is no evidence of a surface melt; under conditions of rapid heating such as burning in a methane atmosphere, a surface melt is observed on the AP; and in the case of self-sustaining deflagration, the surface melt is frothy due to the entrapment of transpiring gases. This frothy melt is probably the site of the major heat release necessary to sustain deflagration.

6.2. BEHAVIOR OF ALUMINUM IN COMPOSITE PROPELLANTS

Current work has indicated that an important step in the aluminum agglomeration (sintering together) process is characterized by the fracture of the aluminum oxide coating (below its melting point) due to stress produced by the differential thermal expansion of aluminum and aluminum oxide as the particles are heated by the combustion wave and oxidative self-heating. Molten aluminum flows through the cracks, joins with adjacent particles and is oxidized, thus forming aluminum/aluminum oxide particles. By including this important step of the agglomeration process, it is possible to give a detailed, qualitative description of the behavior of aluminum in composite propellants from the unheated condition in the propellant to the condition of molten balls of aluminum with oxide "caps" burning by a vapor transport mechanism in the gas phase above the propellant surface.

6.3. NONACOUSTIC COMBUSTION INSTABILITY

Two analytical models of solid propellant combustion instability applicable to the low frequency regime have been reduced to a common notation, were found to be numerically identical, and have been compared with experimental data. Only qualitative agreement between experimental data and model predictions was found, and even then several large

discrepancies were discovered. One discrepancy associated with the heterogeneous nature of the propellant was experimentally explored in detail. A simple, phenomenological argument relating oscillation frequency to oxidizer particle size was developed and, when applied in conjunction with the analytical models, was found to be useful in interpreting the experimental data.

6.4. ACOUSTIC INSTABILITY

Experiments with pressed AP-Al material in the NWC 5.5-inch T-burner indicate that such material can be successfully used to investigate the interaction of acoustic waves with spontaneously occurring combustion oscillations.

Investigation of nonisentropic behavior has shown that phenomena were involved which were not considered in presently available mathematical models. Experimental information acquired during this report period indicated that flame temperature variations and gas velocity perturbations were of the same order of importance as the temperature fluctuations induced by variations in entropy. Steps were initiated to synthesize a more relevant mathematical model which will include consideration of nonuniform gas velocity and a variable flame temperature, as well as improvements in the methods of obtaining experimental measurements.

Appendix A

BURNING RATE DATA AND TABULAR EXPERIMENTAL DATA OBTAINED
WITH RESPECT TO NONACOUSTIC COMBUSTION INSTABILITY

TABLE A-1. Data Obtained With A-146 Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} , msec	Dimensionless frequency $\frac{a_f \omega}{r^2}$	$\omega \tau_c$	Phase shift radians	Real part of the response function
906	20	168	168	.162850	7.3	.897
	32	55	195	.168	1.360	2.2	.469
907	18	162	145	.154765	7.8	.777
908	44.8	158	.159	1.904
	15.9	165	141	.153	97.3	.676	8.0	.699	.580	1.07
	17.1	150	148	.155	114	.727	7.1	.684	.563	1.08
	19	131	153	.157	39.1	.808	6.1	.661	.570	1.03
	21	131	153	.157	-37.8	.893	6.1	.731	.647	.936
910	14.6	172	121	.144	113	.621	9.5	.669	.559	1.07
	16.1	150	130	.148	54.3	.684	8.3	.679	.578	1.04
	18.5	133	131	.149	36.7	.786	6.8	.655	.567	1.03
	24.2	104	130	.148	1.029	5.4	.671
899	45	56	95	.129	27.2	1.913	3.8	.671	.568	1.05
901	13.8	171	115	.141587	9.8	.629
	15.1	165	125	.1465	55.5	.642	8.8	.665	.571	1.04
	16.8	133	125	.1465	70	.714	7.1	.594	.515	1.05
912	14.0	158	120	.143	80.5	.595	8.9	.590	.513	1.05
	15.1	148	125	.1465	85	.642	7.9	.595	.513	1.05
	16.5	139	127	.147	61.8	.701	7.3	.612	.531	1.04
	18.9	127	124	.146	38.5	.803	6.8	.642	.557	1.03
	23.5	105	121	.144999	5.8	.659
	28.6	97	122	.145	1.216	5.3	.739
914	12.4	160	98	.131	139	.527	10.7	.529	.458	1.07
	13.0	162	105	.135	126	.553	10.2	.562	.483	1.07
915	14.1	148	102	.133	95	.599	9.6	.555	.483	1.06
	15.8	128	106	.136	96	.672	7.9	.539	.468	1.07
916	23.6	100	93	.127	67	1.003	7.1	.628	.532	1.07
	26.2	98	101	.133	53.3	1.114	6.3	.683	.573	1.06
917	29.3	83	98	.131	*45.8	1.245	5.5	.648	.550	1.06
918	39.2	72	96	.130	-17.5	1.666	4.875	.753	.660	0.97
	42.4	58	85	.122	- 5.3	1.802	4.450	.655	.584	1.00

TABLE A-1. (Contd.).

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} , msec	Dimensionless frequency $\frac{a_f \omega}{-2}$	$\omega \tau_c$	Phase shift radians	Real part of the response function
919	.6.9	117	101	.133	59.2	.718	7.573	.527	.468	1.04
	18.6	111	106	.136	48.8	.791	6.875	.550	.487	1.04
	20.6	102	104	.134	67.4	.876	6.50	.559	.486	1.06
920	15.0	140	90	.126	51	.638	10.08	.560	.497	1.03
	20.5	127	97	.130	85	.871	8.60	.695	.574	1.07
	23.7	128	100	.132	46.8	1.007	8.40	.810	.659	1.05
921	28.5	95	101	.133	91.8	1.211	6.15	.723	.577	1.12
922	23.5	91	95	.129	51.5	.999	6.25	.570	.497	1.05
	26.2	84	97	.130	54.7	1.114	5.675	.586	.504	1.06
923	27.9	75	88	.124	33.6	1.186	5.575	.558	.493	1.04
	31.8	79	95	.129	35.5	1.352	5.425	.668	.568	1.10
	36.5	65	92	.127	31.9	1.551	5.25	.720	.599	1.05
925	46.4	52	87	.1235	1.972	3.90	.643
	50.7	44	90	.126	2.155	3.175	.596
	54.1	46	90	.126	2.299	3.3	.660
	58.3	42	83	.120	20.7	2.478	3.325	.650	.554	1.05
	58.3	42	83	.120	-18.2	2.478	3.325	.650	.597	.95
927	23.0	80	84	.121	39.8	.978	6.25	.490	.441	1.04
	24.3	80	90	.126	45.5	1.033	5.75	.522	.462	1.05
928	27.2	65	81	.1185	57.2	1.156	5.30	.471	.416	1.06
	34.9	66	87	.1235	32.6	1.483	4.95	.614	.530	1.05
929	45.3	49	84	.121	18.7	1.925	3.825	.591	.519	1.04
	45.3	49	84	.121	-17.7	1.925	3.825	.591	.549	0.98
930	73.8	34.5	76	.114	-18.2	3.137	3.025	.675	.621	0.94
956	~35	63	70	.109	1.49	6.075	.588
957	~53	48	65	.104	2.25	5.075	.686
958	~85	63	.1025
960	~48	45	60	.0995	2.04	5.2	.576
	~55	33	62	.1015	2.34	3.675	.485
962	~81	28	65	.104	3.44	2.950	.602

TABLE A-2. Data Obtained With A-148 Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psio	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} , msec	Dimensionless frequency $\frac{\alpha_1 \omega}{-2}$	$\omega \tau_c$	Real part of the response function
1001	19	150	114	.116806	13.2	.759
	20.1	125	114	.116855	11.0	.671
	23.2	127	115	.117	66.2	.98	11.2	.781	1.07
	23.8	119	113	.116	57.2	1.01	10.5	.754	1.06
	24.9	116	111	.115	36.0	1.06	10.4	.772	1.04
	27.2	108	108	.115	22.3	1.16	9.76	.786	1.03
	31.3	100	105	.113	0	1.33	9.23	.835	1.00
	32.7	98	104	.113	0	1.39	9.04	.855	1.00
	40.2	73	90	.107	28.7	1.71	7.56	.783	1.05
1002	25	100	81	.102	1.06	11.4	.666
	25.6	~100	85	.105	1.09	11.0	.686
	26.5	97	85	.105	1.13	10.7	.688
	27.1	87	82	.103	1.15	9.39	.628
	28.2	91	84.5	.105	1.20	9.99	.685
	28.5	89	84	.104	1.26	9.78	.704
	30.6	85	86	.106	1.30	8.70	.694
	32.2	80	83	.103	1.37	8.48	.688
	33.6	81	80	.102	1.43	9.47	.728
	35.0	80	80	.102	1.49	9.34	.748
1003	33	72	70	.0955	1.42	9.49	.642
	36.6	70	.0955	1.56
	40.2	65	.0915	34.3	1.71	1.06
	45	57	67	.0935	27.2	1.91	7.70	.684	1.05
1124	17.5	115	91	.10774	11.5	.534
	18.2	120	94	.10878	11.8	.588
	19	110	93	.10881	10.8	.560
	19.7	120	93	.10883	11.8	.626
	20.7	94	.10888
	22	94	.10894
1125	28.1	80	82	.104	1.20	8.48	.602
	29.8	80	94	.108	84.0	1.27	7.83	.638	1.11
	33.8	87	97	.109	29.2	1.43	8.35	.781	1.04
	40.6	75	94	.108	25.8	1.73	7.35	.815	1.05
1126	44.7	71	94	.108	19.0	1.90	6.96	.847	1.04
1127	19	100	103.5	.113	28.2	.809	8.92	.508	1.02
	22	110	107.5	.113	5.2	.94	9.81	.650	1.01
	25.9	105	105.0	.112	-16.2	1.10	9.56	.725	.98
1128	18.3	115	107	.11578	9.89	.563
	20.4	133	117	.117	76.7	.87	11.1	.726	1.07
	25.3	~ 6.7	120	.118	0	1.01	.55	.045	1.00
1129	50.7	72	93.5	.110	2.16	6.78	.976

TABLE A-2. (Contd.).

Run no.	L^* , in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec^{-1}	τ_{ch} , msec	Dimensionless frequency $\frac{\alpha_1 \omega}{\tau^2}$	$\omega \tau_c$	Real part of the response function
A-148A (different burning rate)									
1215	32-39	~ 5.8	265	~.17	~ 0	1.36-1.66	.23	.050 .060	~1.0
1216	28	~16	270	~.17	~ 0	1.19	.63	.119	~1.0
1217	18.0	170	120	.15	31.0	.76	8.65	.812	1.02
	22.2	143	123	.152	-24.3	.94	7.09	.844	.98
	35.2	18.5	120	.15	low amp.	1.50	.94	.174
	~45	62	92	.135		1.91	3.89	.743
1218	14.1	140	94	.135	+	.60	8.79	.527
	14.5	179	109	.144		.62	9.89	.697
	15.7	175	108	.143	~81	.67	9.78	.736	~1.05
	18.1	135	111	.145	~76	.77	7.38	.653	~1.06
	20.7	133	110	.145	~70	.88	7.26	.735	~1.06
	24.1	134	106	.142	~58	1.02	7.58	.859	~1.06
	27.0	117	101	.139	~65	1.15	6.91	.845	~1.08
	30.3	108	96	.137	~57.5	1.29	6.58	.875	~1.07
	33.7	100	93	.135	1.43	6.28	.898
1219	29	133	116	.147	~ 0	1.23	7.01	1.027	1.00
	37	~ 9.5	108	.143	low amp.	1.57	~.53	~.094
	45	72	94	.135	28.2	1.91	4.53	.865	1.05
	48.7	62	84	.128	41.5	2.07	4.36	.814	1.06
1220	50.1	60	76	.121	~68.5	2.13	4.67	.803	1.15
1221	53.4	47	68	.115	2.27	4.04	.670
	57.1	46	71	.117	33.7	2.43	3.84	.702	1.08
	60.5	50	65	.112	44.0	2.57	4.55	.807	1.11
1222	38.5	42	77	.121	~33.0	1.64	3.24	.428	~1.05
1225	28.6	101	87	.130	51.0	1.22	6.86	.775	1.06
	35.5	93	86	.129	46.0	1.51	6.37	.882	1.07
	44.4	91	88	.130	47.5	1.89	6.17	1.079	1.09

TABLE A-3. Data Obtained With A-149 Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} msec	Dimensionless frequency $\frac{a_g \omega}{-2}$	$\omega \tau_c$	Real part of the response function
1016	106.3	10.8	68	.064	4.51	3.0	.306
	116	10.7	69	.064	4.93	3.0	.332
1019	41	30	130	.084	1.74	4.86	.328
	54.5	23.4	150	.089	2.32	3.38	.341
	61.4	20	158	.091	2.61	2.77	.328
	64.4	27.3	158	.091	2.74	3.78	.470
1020	57.5-	151.5	.089	2.42-
	107					4.52			
1021	35	18.8	108	.077	1.49	3.64	.176
	37.7	25	110	.078	1.60	4.7	.251
1022	60	20	103	.075	2.55	4.07	.321
1023	70.3	13.3	55	.058	5.42	2.99	4.5	.249	1.04
	89	13.2	58	.059	-4.46	3.78	4.45	.314	.98

TABLE A-4. Data Obtained A-151 Propellant.

Run no.	L^* , in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ , msec	Dimensionless frequency $\frac{\omega \tau}{\sqrt{2}}$	$\omega \tau_c$	Real part of the response function
1074	56.5	72	101.5	.118	50	1.55	5.9	.700	1.08
1078	21.9	77.0	77	.105	40.8	1.55	8.0	.650	1.06
	29.4	72.0	76.5	.105	1.68	7.5	.770
1080	35.0	47.6	75.0	.104	48.2	1.48	6.60	.442	1.07
1100	24.5	34.5	65.0	.097	1.04	6.60	.356
	34.9	50	68.5	.099	40.9	1.48	5.82	.465	1.05
1101	45.3	40	61.5	.093	1.92	5.3	.482
	51.3	37	63.5	.095	24.5	2.18	4.65	.507	1.05
	57.5	56.5	58.5	.090	58.6	2.44	5.15	.560	1.07
1103	69.7	33.5	58.5	.090	41.6	2.96	4.7	.620	1.12
	78.3	28.5	57.5	.090	21.2	3.33	4.0	.596	1.07
1104	93.2	22	53.5	.086	19.1	3.96	3.94	.548	1.08
1107	14.2	50	75	.104	0.60	5.29	.188
	15.7	55	76	.105	0.66	5.72	.228
	17.0	50	77	.105	0.72	5.19	.226	1.02
	23.4	44	73.5	.103	23.8	0.99	4.74	.274
	34.9	42	71.0	.100	12.8	1.48	4.90	.400	1.00

1115	44.1	40	70.3	.100	~0	1.87	4.56	.470
	50.4	38	68.5	.099	~0	2.14	4.45	.511
	59.3	51.5	65.5	.097	17.0	2.52	3.82	.499	1.04
1117	29.2	50	63.5	.099	23.5	1.24	5.83	.590	1.04
	35.2	35	66.5	.099	34	1.41	6.43	.487	1.07
	41	50	70.3	.103	50	1.74	5.4	.547	1.09
1132	19.6	80	106	.125	~0	0.83	5.85	.417
	44.4	40	72.5	.104	16.8	1.88	4.22	.473	1.02
1133	18.5	66.5	94.0	.120	36.3	0.78	5.28	.334	1.02
	23.1	11.8	97	.120	~0	0.98	0.94	.0724
	24.3	70	95	.119	~0	1.03	5.65	.453
1135	44.9	36.1	54.5	.089	1.91	5.30	.433
	50.8	36	56.5	.090	25.5	2.16	5.08	.488	1.05
1142	47	15.5	~0.030	59.6
1144	25	40	71.5	.105	15.4	1.06	4.32	.267	1.02
	29.5	45	68.5	.100	1.56	5.13	.356
1145	34.2	44.6	57.5	.093	40	1.45	5.87	.404	1.04
	38.1	42.8	59.5	.094	14.6	1.62	5.55	.436	1.02
1144	59.6	27.2	52.5	.088	14.2	2.34	3.84	.434	1.04
	55.6	18.5	~0.036	49.2
1134	60.8	29.5	51.0	.086	2.58	4.55	.478
1172	61	31	47.5	.075	2.58	4.28	.502
1173	85	25	45	.072	3.60	5.50	.566
1178	16.5	80	86	.104	23	0.70	8.12	.352	1.02
	18.2	70	105	.125	~0	0.77	5.12	.339
	24.6	15	117	.133	~0	1.04	0.97	.098
1179	20.3	67	75.5	.099	0.86	7.80	.363
	21.3	59	73.5	.098	0.90	7.05	.334
	25.7	53.5	80	.102	8.8	1.09	5.88	.364	1.01
	27.6	48.5	85.5	.105	-19.4	1.17	5.05	.354	1.00
	42.2	43	76.0	.100	18.5	1.79	4.93	.484	1.03
	46.5	62	33.5	.060	1.97	19.67	.748

TABLE A-5. Data Obtained With A-155 Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{chr} , msec	Dimensionless frequency $\frac{\alpha_1 \omega}{\tau^2}$	$\omega \tau_c$	Real part of the response function
1165	18.2	83	87.5	.092	57.6	.772	11.72	.402	1.04
	20.8	62	83.5	.090	38.4	.885	9.15	.344	1.03
	21.7	57	80.5	.087	-21.2	.935	8.60	.331	.98
	24.5	50	90.5	.093	8.6	1.04	6.62	.327	1.01
	26.6	50	85.0	.090	1.13	6.12	.355
	28.6	50	88.0	.092	- 8.8	1.22	6.9	.383	.99
	45.6	42	79.5	.087	21.8	1.94	6.50	.512	1.04
1166	31.2	40	76.5	.085	27.5	1.32	6.48	.331	1.04
	40.6	41.5	73.5	.092	36.0	1.73	7.08	.451	1.06
	47.6	37.0	73.5	.082	24.1	2.02	6.30	.469	1.05
1167	69.1	30.0	68.5	.079	16.1	2.94	5.32	.553	1.05
	75.8	29.8	62.5	.075	3.22	6.06	.602
1168	53.7	32.3	72.5	.082	16.1	2.28	5.5	.463	1.04
	65.3	30.8	67.5	.079	22.7	2.78	5.36	.537	1.06
1170	33.8	40	83.5	.090	-24.7	1.43	5.91	.359	.97
	59.5	29	75.0	.084	28.3	2.53	4.82	.460	1.07
1188	82	33.3	78.5	.086	3.48	5.14	.727
	96.4	33.3	77.5	.085	4.1	5.26	.857
	101.6	32	78.5	.086	4.33	4.95	.870
1189	58.2	29.0	81.5	.087	2.47	4.37	.450
	66.0	26.7	77.5	.085	29.3	2.80	4.25	.470	1.07
	69.4	25.3	80.5	.087	2.94	3.80	.467
1190	87.4	22	80.5	.087	3.72	3.32	.513
1191	112	22	71	.081	4.75	3.82	.656
1203	23.4	55.6	109.0	.105	1.02	5.76	.356
	43.5	3.9	110.5	.106	1.85	.39	.045
1205	72	28.5	67.5	.079	3.06	5.25	.542
	76	26.8	64.0	.076	26.0	3.24	5.30	.544	1.08
1206	112	20	53.5	.069	-15.3	4.67	4.80	.588	.93
	118	19.6	53.5	.069	26.2	4.92	4.71	.605	1.13
	125	20	58.5	.073	5.3	3.13	.668
1226	24.1	52	92	.096	-54.8	1.025	6.45	.335	.94
	39.4	2.67	100.5	.101	1.67	.30	.028
1232	20.8	56.0	83.5	.090	-41	.885	7.90	.312	.96
	24.7	55.0	88.0	.094	-32	1.05	7.10	.370	.96
	31.0	54.5	91.5	.095	1.32	6.90	.451
	45.9	3.0	97.5	.099	1.95	.35	.037
	53.0	34.9	86.2	.092	2.25	4.7	.493

TABLE A-6. Data Obtained With A-156A Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} msec	Dimensionless frequency $\frac{\omega_p \omega}{\tau^2}$	$\omega \tau_c$	Real part of the response function
1227	55	15	175	.144	2.34	.825	0.220
1229	50.8	40	64	.105	2.16	4.15	.543
1238	25	115	68.5	.108	71.5	1.06	11.3	.766	1.08
	35	100	73.5	.109	35.0	1.49	9.65	.936	1.05
1241	30.4	71	57.5	.102	1.29	7.80	.575
	32.5	77	58.5	.102	1.38	8.45	.667
	34.0	67	57.0	.100	1.44	7.65	.606
	34.7	67	57.5	.101	1.47	7.50	.619
	35.4	71	58.5	.102	1.50	7.80	.669
1242	36	85	34	.072	1.53	18.8	.817
1243	55	64	34	.072	2.34	14.2	.941
1254	120	13	152	.140	~ 0	5.1	.755	.416	1.0
1255	86- 124	189	.145	3.66- 5.27
1256	159	14	104	.123	6.76	1.06	.594
1257	19	97	116	.127808	5.05	.492
	20- 33	14	120	.128850- 1.40	.975	.075- .124
1258	42	118	.127	1.79
1259	40.3	42.3	73	.108	22.6	1.71	4.15	.454	1.04
	44.0	43.5	70	.107	-13.7	1.87	4.35	.511	0.97
	46.4	44.5	69	.107	~ 0	1.97	4.45	.551	1.0
1260	56- 75	80	.110	2.38- 3.19
1262	15.7	50.0	76	.109	+	.57	4.80	.210
	16.1	50.6	76	.109	~ 0	.68- .80	4.85	.216- .254	1.0
	20.7	40.0	71	.108	9.9	.88	3.92	.221	1.01
	21- 25	45.5	72	.108	22.6	.89- 1.06	4.45	.254- .303	1.02
1263	19.3	95	108	.125	46.4	.82	6.95	.489	1.04
	43.4	57	85	.111	27.8	1.85	5.30	.642	1.05
	44.5	43.5	78	.110	~ 0	1.89	4.11	.516	1.00
1264	18.0	85.6	110	.12577	6.27	.414
	19.0	88.4	109	.12581	6.43	.450
	20.0	83.5	110	.12585	6.11	.446
1265	73.7	32.2	60	.101	3.13	3.62	.633
	87	29.4	66	.106	3.70	3.00	.683

TABLE A-6. (Contd.).

Run no.	L*, in	Frequency, cps	Pressure, psia	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} , msec	Dimensionless frequency $\frac{\alpha_1 \omega}{-2}$	$\omega \tau_c$	Real part of the response function
1266	90.1	14.3	66	.106	3.83	1.46	.344
	94.0	14.3	66	.100	4.00	1.46	.359
	104.0	19	66	.106	4.42	1.94	.527
	107.6	15.4	61	.102	4.57	1.69	.442
1267	26	73.5	94	.116	27.6	1.10	6.25	.508	1.03
	27.6	60.4	90	.115	40.0	1.17	5.23	.444	1.05
1268	51.5	25.6	61	.101	0	2.19	2.87	.352	1.00
	55.4	40.0	74	.108	22.5	2.35	3.94	.590	1.05
1269	80.9	42.9	78	.109	3.44	4.12	.927
1270	17	92	106	.12472	6.84	.416
	19	87.6	105	.123	28.6	.81	6.62	.446	1.02
1270B	37	13.4	114	.126	1.57	.965	.132
	40	16.6	119	.128	1.70	1.16	.177
	44	14.3	141	.136	1.87	.885	.168

TABLE A-7. Data Obtained With A-157 Propellant.

Run no.	L*, in	Frequency, cps	Pressure, psio	Burning rate in/sec	Growth constant, sec ⁻¹	τ_{ch} , msec	Dimensionless frequency $\frac{\omega \tau_c}{\sqrt{2}}$	$\omega \tau_c$	Real part of the response function
1159	31.9	46	76	.117	27.3	1.36	3.84	.393	1.037
1161	53	33	69.5	.110	2.25	3.12	.466
1163	24.1	62.5	73.5	.115	1.02	5.40	.401
	25.9	70.0	83.5	.120	1.10	5.56	.484
1194	30	47	93.5	.120	31.5	1.28	3.73	.378	1.04
1197	27.1	51.5	88.5	.120	31.8	1.15	4.09	.372	1.04
	30.0	55.0	88.5	.120	32.0	1.28	4.36	.443	1.04
	31.6	61	90.5	.120	1.34	4.85	.514
	34.9	50	87.0	.120	38.4	1.48	3.97	.465	1.06
	39.0	50	83.5	.120	37.0	1.66	3.97	.523	1.06
1198	45.6	22.8	63.0	.103	1.94	2.46	.278
1200	68	27.5	63.5	.103	2.89	2.97	.499
	73	33.3	60.5	.101	3.10	3.72	.649
1202	66.1	36.0	72.5	.114	23.4	2.81	3.16	.635	1.07
	74.1	35.2	75.5	.115	3.15	3.04	.696
	79.3	31.0	58.5	.095	3.37	3.94	.656
1211	20.1	85	98.5	.12085	6.75	.454
	27.0	75	96	.120	1.15	5.96	.542
1212	17.6	82	90.5	.120	17.2	.75	6.51	.388	1.01
	26.2	80	90.5	.120	65.2	1.11	6.35	.558	1.07
1213	29	83.5	82	.120	79	1.23	6.64	.645	1.10
1214	70	33.3	41.5	.078	2.98	6.25	.624
1235	16.6	85	85.5	.12071	6.75	.378
	17.6	68	86.0	.12075	5.40	.321
	18.7	66	85.5	.120	51.0	.79	5.25	.425	1.04
	20.0	67	84.0	.120	41.8	.85	5.32	.358	1.04
	21.1	65	84.0	.120	48.8	.90	5.16	.368	1.04
	22.9	63	83.5	.120	46.8	.97	5.00	.384	1.05
	24.4	55.5	84.0	.120	51.5	1.04	4.41	.363	1.03
	25.4	61	83.0	.120	46.0	1.08	4.85	.414	1.05
	29.7	52.5	83.0	.120	31.2	1.26	4.17	.415	1.04
	31.6	61.5	88.0	.120	1.34	4.88	.517
	34.9	59.0	85.5	.120	1.48	4.68	.549
	36.6	55.5	84.0	.120	1.56	4.41	.543
	38.6	48.5	82.0	.120	1.64	3.86	.500
1237	26.6	60.0	77.5	.119	1.13	4.85	.426
	27.3	61.0	78.5	.119	1.16	4.93	.444
	28.0	67.5	78.5	.119	1.19	5.45	.505
	29.6	57	79.0	.119	1.26	4.60	.452
	31.1	50.0	77.5	.117	48.6	1.32	4.17	.415	1.06
	32.3	53.5	73.5	.115	23.2	1.37	4.64	.461	1.03
	34.0	53.2	78.5	.119	- 9.7	1.44	4.30	.481	.99
	36.6	48.9	78.5	.119	37.2	1.56	3.94	.480	1.06
	39.5	46.2	80.0	.120	32.9	1.68	3.66	.488	1.06
	43.6	46.3	76.0	.117	28.8	1.85	3.87	.537	1.05
	45.4	49.2	73.0	.114	1.93	4.31	.596

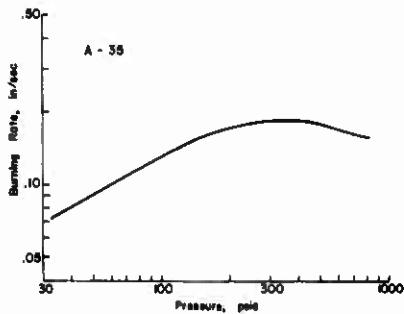


FIG. A-1. Burning Rate for Propellant A-35.

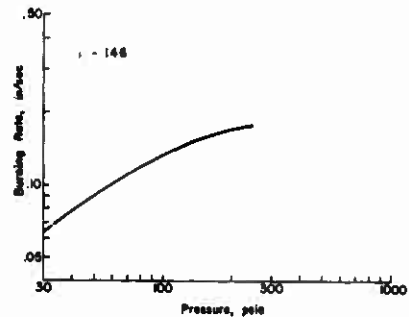


FIG. A-2. Burning Rate for Propellant A-146.

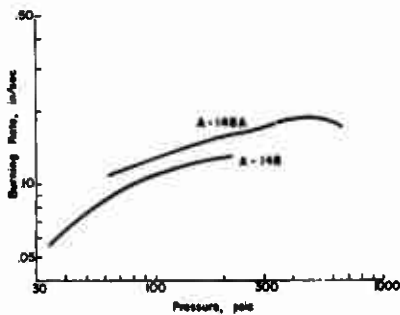


FIG. A-3. Burning Rate for Propellant A-148.

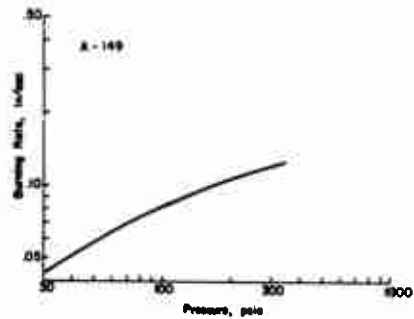


FIG. A-4. Burning Rate for Propellant A-149.

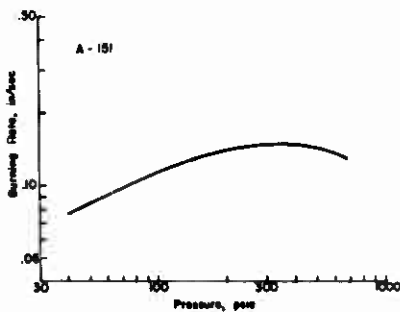


FIG. A-5. Burning Rate for Propellant A-181.

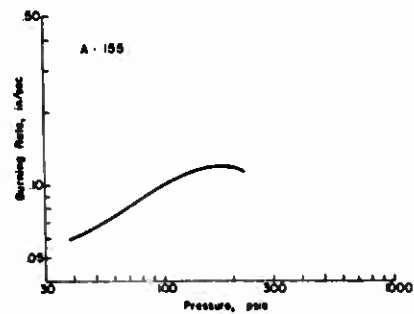


FIG. A-6. Burning Rate for Propellant A-155.

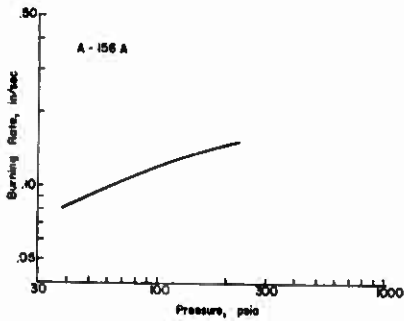


FIG. A-7. Burning Rate for Propellant A-156.

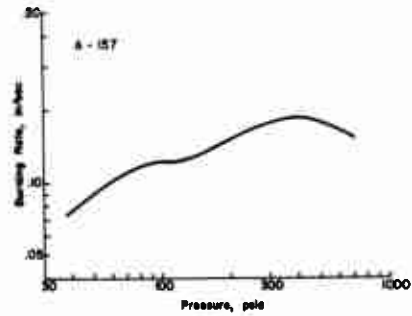


FIG. A-8. Burning Rate for Propellant A-157.

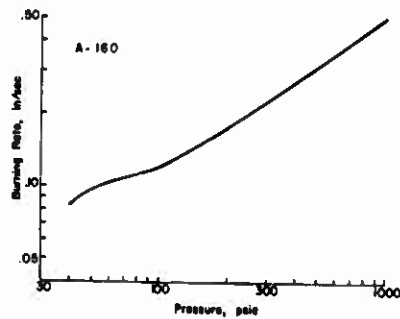


FIG. A-9. Burning Rate for Propellant A-160.

Appendix B

INSTABILITY TESTS ON CANDIDATE
BIG BOOSTER PROPELLANTS

Two propellants that had been designed for and were candidates for use in the large solid booster program were tested in both the L*-burner and the low-frequency T-burner. They were not tested in the conventional T-burner because of the high metal loading in the propellants which would cause high-frequency oscillations to be completely attenuated.

Testing in the L* burner was more extensive than that of the "big pipe" due mainly to the ease of testing and the smaller amount of propellant used in the L*-burner. Instability was observed for both propellants with the bounds that are normally observed in this burner. The propellants both behaved in what would be termed a "normal" fashion and on this basis are considered as being typical, i.e., nothing occurred that would indicate that they would be more prone to instability than any other propellant. The data that were obtained are included in Tables B-1 and B-2.

TABLE B-1. Data Obtained With ANB-3105

Run no.	L*, in.	Frequency, cps	Pressure, psia	Growth constant, sec ⁻¹
1008	11.4	59	80	< 0
1009	7.8	58	66	> 0
	8.7	53	70	~ 0
1010	13.6	44	48	> 0
	15.2	46	50	> 0
	16.7	44	55	< 0
1011	25.1	39	52	~ 0
1012	25.1	37	50	~ 0
	26.2	37	57	~ 0
	27.9	39	67	~ 0
1013	17.8	43	48.5	~ 0
	22.3	45	56.5	~ 0
1014	8.86	52	71.0	19.8
	9.62	52	70.5	~ 0
	10.4	51.6	70.0	-12.7
	16.56	53.0	83.5	~ 0

TABLE B-1. (Contd.)

Run no.	L*, in.	Frequency, cps	Pressure, psis	Growth constant, sec ⁻¹
1047	10.56	65.5	46	12.8
	11.41	65.0	50.5	- 7.52
1048	7.7	75	73.5
	8.0	68	70.5	22.3
	8.4	66.7	73.4	9.5
	9.1	60.0	73.2	17.7
	10.5	70	80.5	- 0
1049	13.9	77.3	80.0	11.1
	14.3	68.0	78.5	25.6
	14.9	62.0	76.5	24.1
	16.1	60	70.0	14.7
	18.8	58	72.5	- 0
1050	6.1	55.5	57.5	17.6
	7.5	54.5	67.0	-21.9
	8.4	58.0	71.5	- 0
1051	11.2	80.0	69.5
	12.1	70.0	60.5	28.0
	13.0	66.7	56.5
	14.2	55	50.5	50.5
	15.8	50.0	47.5	32.9
	16.4	50.0	43.4
	17.7	47.6	42.5	48.8
1052	26.3	40	39.5	32.3
	28.2	40	40.5	18.8
1053	35.7	33	47.5	7.0
	36.8	36.4	46.5	8.1
	40.1	31.0	38.0	23.2
	41.5	32.1	35.0	14.5
1054	20.3	22.0	25.5	21.0
	21.3	25.0	29.5	18.5
	22.4	26.2	33.4	22.1
	25.0	29.2	37.5
1090	21.3	60	71.5	22.6
	9.7	62.5	71.5	-22.9
1092	11-31	160
1093	10	62	55	- 0
1094	14.7	40	46.0	12.8
1095	20-33	112

TABLE B-2. Data Obtained With ANB-3254 Propellant

Run no.	L*, in.	Frequency, cps	Pressure, psia	Growth constant sec ⁻¹
1039	22.4	50	60.5	70
	21.8	54	78.5	0
1061	87
1062	98
1070	35	35.6	70.5	24.6
	100.5
1072	38.5	30	50.5	-14.7
1073	47.5	33	67	-13.5
1081	27.8	40	67.5
	26.4	37.5	70.5	-16.3
	30.7	31	62.5	29.5
1082	~35	41	67	0
1083	~69	30	57.5	-10.9
1084	7.6	65	92.5	-25.7
1085	~12	55	68.5	-21.8
1086	- 7.2	75	105	33.0
1087	7.6	75	95	-29.0
1088	20.9	50	73.5	32.4
1089	~27	43	83.5	-18.2

LOW FREQUENCY ACOUSTIC TESTS

Seven tests were conducted in the 5.5-inch T-burner ranging from 15 to 250 cps. The pressure control system was programmed to provide for a progressively increasing pressure during burning of the propellant. The high burning rate of these two propellants resulted in a tendency for the relatively great volume of combustion products to override the pressure control system and to create a burner pressure-time history that deviated considerably from that which was desired. No oscillatory behavior was observed which could be assigned to combustion-driven processes, except in one test with ANB-3254. Some mild oscillations were observed in other tests which appeared to be caused by vibration of pressure transducers or induced by the pressure control apparatus. Test information is detailed in Table B-3.

CONCLUSIONS

As a result of testing these two propellants, it was concluded that the possibility of either of these propellants experiencing pressure coupled acoustic instability or nonacoustic instability in the 260-inch motor configuration would be rather remote. The possibility of velocity

coupled instability was not tested for and could not be ruled out. However, although the possibility of this type of instability occurring exists, it was considered unlikely in the 260-inch configuration.

TABLE B-3. 5.5-inch T-Burner Tests

Propellant	Test no.	Burner length, ft	Pressure range, psig	Pressure range of Osc. psig	Freq.	Remarks
ANB 3105	4820	12	20-63	...	→	Low amplitude signal ~ 250 cps at end of burn
	4776	24	24-70	...	→	Low amplitude 300 cps signal above 25 psig
	4777	24	0-35	...	→	(May be transducer acting up)
ANB 3254	4821	6	0-72	55-72	250	Small amount of higher frequency present
	4819	12	40-90	...	Stable	Poor pressure control
	4778	24	30-63	...	Stable	Poor pressure control
	4818	48	0-55	...	Stable	2nd mode in last portion of test-probably due to air controller

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